



Research paper

Occurrence and behaviour of uranium in the groundwater and potential health risk associated in semi-arid region of Punjab, India

Ritu Bala, Karanveer, Debabrata Das^{*}

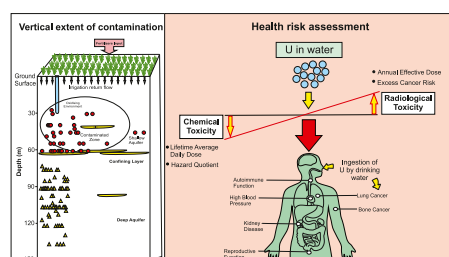
Department of Geology, Panjab University, Chandigarh, 160014, India



HIGHLIGHTS

- Shallow alluvial aquifer is more contaminated with high spatial variability.
- U levels are higher along the groundwater flow direction in the SW direction.
- EC, Na⁺, K⁺ and HCO₃⁻ ions are primarily governing the behaviour of uranium.
- Weathering of carbonates is a major cause of uranium mobilization in the aquifers.
- 33% of samples exhibit carcinogenic risk due to U in groundwater.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Groundwater
Uranium
Stable isotope
Human health risk
Punjab

ABSTRACT

Occurrence of uranium (U) in the groundwater of south-west Punjab is a matter of great concern due to the rise of contaminant levels at an alarming rate, its spatial and vertical variability and poor understanding of uranium mobility with depth. Present study has been conducted to investigate the vertical extent of contamination and geochemical factors controlling U mobilization in the groundwater of semi-arid Punjab. Out of total 140 samples collected and analyzed, 76% samples have uranium levels greater than the chemical toxicity limit of World Health Organization (WHO, 30 $\mu\text{g.L}^{-1}$) and 34% samples have concentration higher than the radiological toxicity limit given by Atomic Energy Regulatory Board (AERB, 60 $\mu\text{g.L}^{-1}$). Water at shallow depth (<60 m) is found to be more contaminated than water at deeper depth (>60 m) and concentration lies in the range of 4.26–318.03 $\mu\text{g.L}^{-1}$. Spatial distribution graphs are used to identify the hotspots of contamination and concentration is higher in the flow direction of groundwater towards south-west. In both the aquifers, groundwater is alkaline and oxic in character with carbonate weathering as dominant hydrogeochemical process affecting chemical composition of water. Higher uranium concentration is seen in the oxidizing, alkaline and carbonate-rich water. Alkali earth metals (Na⁺ & K⁺) are also strongly correlated with the uranium in groundwater at both the levels. Use of environmental stable isotopes ($\delta^{18}\text{O}$ & $\delta^2\text{H}$) shows the evaporation signature in both the aquifers. Health risk assessment is carried out and values higher than permissible limit give indication towards the health risk to the population exposed due to the consumption of uranium contaminated water from long time. This study provides

Abbreviations: AERB, Atomic Energy Regulatory Board of India; BDL, Below Detection Limit; Ca²⁺, Calcium; Cl⁻, Chloride; cm, centimeter; EC, Electrical conductivity; HCO₃⁻, Bicarbonate; ²H, Deuterium; K⁺, Potassium; m, meter; mg.L⁻¹, milligram per litre; mg.kg⁻¹, milligram per kilogram; Mg²⁺, Magnesium; Na⁺, Sodium; NO₃⁻, Nitrate; NW, North-west; ¹⁸O, Oxygen-18; ORP, Oxidation-reduction potential; PO₄³⁻, Phosphate; SO₄²⁻, Sulphate; SW, South-west; TDS, Total Dissolved Solids; U, Total Uranium; WHO, World Health Organization; $\mu\text{g.L}^{-1}$, microgram per litre.

^{*} Corresponding author.

E-mail address: debabratadas@pu.ac.in (D. Das).

<https://doi.org/10.1016/j.gsd.2022.100731>

Received 21 September 2021; Received in revised form 7 January 2022; Accepted 23 January 2022

Available online 29 January 2022

2352-801X/© 2022 Elsevier B.V. All rights reserved.

a strong base for better understanding the source of uranium in the aquifer system of region and the results would be useful for further studies in quaternary alluvial aquifers of semi-arid regions.

1. Introduction

Uranium (U) as a potential drinking water health hazard is brought to the notice after the provisional guideline given by World Health Organization (WHO). Earlier U was not a part of water quality investigations at most of the places and thus the background information on distribution and range of concentration in the groundwater is limited. From past few decades, element concentration in groundwater exceeding the permissible limit of WHO i.e. $30 \mu\text{g.L}^{-1}$ (WHO, 2011a) has become a serious health concern globally. Countries such as Canada (Moss et al., 1983), Finland (Kurtio et al., 2006), New Mexico, USA (Hakonson-Hayes et al., 2002), China (Wu et al., 2014) and Mongolia (Nriagu et al., 2012) have identified uranium in its groundwater. Uranium levels in the groundwater of quaternary fluvial aquifer also exceed the German drinking water limit of $10 \mu\text{g.L}^{-1}$ (Banning et al., 2017).

Uranium is introduced to the groundwater system by the natural geochemical processes (geogenic) or anthropogenic activities (Coyte et al., 2018; Smedley et al., 2006). Geogenic source includes the mineral dissolution of host rocks and sediments along the flow path of groundwater or desorption from the surface of various minerals (Chen et al., 2005; Jerden and Sinha, 2006). In rocks and soil, uranium is present in many major rock-forming minerals and is also incorporated in a number of minor phases. It can also be present on grain boundaries. It is well known that clays, carbonate, Fe, Mn, and Al oxides and hydroxides act as strong adsorbent of uranium (Porcelli and Swarzenski, 2003). Anthropogenic sources of uranium include mining and ore processing of uranium, radioactive material production, and disposal, combustion of fuels, and use of phosphatic fertilizers in agricultural fields (Cothorn and Lappenbusch, 1983; Dreeson et al., 1982; Spalding and Sackett, 1972; Tadmor, 1986). However, concentration is highly variable depending on the groundwater chemistry, aquifer characteristics, and bedrock geology (Frengstad et al., 2000).

Redox processes control the enrichment of U in groundwater and its solubility is highly sensitive to the aquifer geochemistry. Fate of the uranium i.e. whether it gets sorbed, mobilized, precipitated or immobilized is decided mainly by its oxidation state and mineral solubility (Maher et al., 2013). Environmental chemistry of uranium is influenced by its oxidation-reduction potential (ORP) because of their redox dependency (Finch and Murakami, 1999). There are three main oxidation states of U(IV, V and VI) in nature, however, U(IV), the reduced form, and U(VI), the oxidized form controls most of the U geochemistry. The solubility of U(VI) is extremely high and oxidizing conditions effectively enhance the uranium movement (Alam and Cheng, 2014). Uranium mobilization is studied by the various complexes formed by it in the water system depending on pH and pCO_2 . At lower pH (<5), dominant species of U is Uranyl ion (UO_2^{2+}), however, UO_2^{2+} forms complexes with carbonate, phosphate, and calcium at higher pHs (Bernhard, 1998; Langmuir, 1978; Porcelli and Swarzenski, 2003).

Uranium concentration is also affected by recharge and discharge from the aquifers and stable isotopes (^{18}O and ^2H) are widely used to get an insight to the aquifer recharge system (Kendall and MacDonnell, 1998; Negrel et al., 2003). These environmental tracers are recognized as recorders of important processes like evaporation, transpiration, recycling, and mixing of water (Tarki et al., 2016). Uranium poisoning is a matter of concern more because of its chemical toxicity than its radiological impacts. A considerable body of evidence also suggests that overexposure to uranium causes nephrotoxicity and osteotoxicity (Kurtio et al., 2006). Uranyl ion replaces the calcium in the bone crystal (Moss et al., 1983) and skeleton is the main site of its accumulation (Wrenn et al., 1985).

In India, 16 states have been detected with uranium in its

groundwater. Uranium in groundwater is prevalent in states of Andhra Pradesh (Babu et al., 2008), Karnataka (Brindha and Elango, 2013), Gujarat, West Bengal, Chhattisgarh, Haryana, Himachal Pradesh, Madhya Pradesh, Punjab, and Rajasthan (Coyte et al., 2018). All the states except north-west (NW) India have Pre-Cambrian rocks which are naturally considered to be a source of uranium (Thivya et al., 2016; CGWB, 2014). The alluvial aquifers of NW India have received worldwide attention from past few decades due to the large scale presence of uranium in its groundwater (Coyte et al., 2018).

Punjab is an agriculture dominant state of NW India and major amount of groundwater is employed for irrigation requirements (Kumar et al., 2018). Aquifers in the region are exploited in terms of both groundwater quantity and quality (Bonsor et al., 2017; Baweja et al., 2017; CGWB, 2017b; MacDonald et al., 2016). Although various studies have been carried out on distribution of uranium in the groundwater of Punjab (Bajwa et al., 2015; Coyte et al., 2018; Pant et al., 2017; Rishi et al., 2017; Singh et al., 1995; Singh and Kishore, 2010; Saini et al., 2016; Sharma et al., 2017), however, work done is very limited in the Barnala district which shows highest groundwater depletion (Sidhu et al., 2021). Current study is important as explained studies on vertical extent of uranium contamination are limited in the region. Due to aforementioned, main objectives of this study are to (a) Know the groundwater quality and vertical extent of uranium contamination (b) link the presence of uranium and water geochemistry by utilizing hydrochemical data (c) understand the source of recharge to the aquifer system by employing environmental stable isotopes and relating it with uranium contamination (d) Study the health risks extent on humans due to uranium ingestion in drinking water. Groundwater being crucial source of water in the region, it is important to understand the levels, source and behaviour of this contaminant. The findings of the current study are going to be useful in understanding the source for future exploration, behaviour of uranium with changing groundwater conditions, for planning remediation and better policy management to reduce the health risk.

2. Material and methods

2.1. Study area

The region forms a part of the Indo-Gangetic alluvial plain and in general, represents a flat topography. It comprises of alluvium and windblown sands. The alluvium is mainly composed of sands of various grades clays and silts and is divided into two parts, newer and older alluvium and is thus heterogeneous in nature in accordance to the mode of deposition by constantly shifting river. Kankar (secondary calcium carbonate) in the form of sheet deposits or in nodular form is met at the depth of 0.75 cm–2 m below the surface. Small sand dunes of varying dimensions and different alignments are observed at a number of places. These dunes can be broadly classified to be of scattered type and do not appear to have any regional linear trend although in isolate parts they do have orientation (CGWB, 2017b).

Study area lies in the south part of Satluj River in the Malwa region of Punjab and lies in the arid & semi-arid climatic zone (Paul et al., 2015). It includes Barnala district which is bounded by Moga and Ludhiana in the north, Bathinda in the west, Mansa in the south and Sangrur district in the east (Fig. 1). Soil of the region is highly fertile and is under extensive agriculture. Land use map of the region has been provided in Fig. 1 (National Remote Sensing Centre, 2015–16). The climate of the area is described as semi-arid, tropical steppe, and hot. Area receives unevenly distributed normal monsoon and average rainfall of 434 mm and 504 mm respectively (CGWB, 2017a).

The region forms part of both Ghaggar and the Satluj river sub-basin.

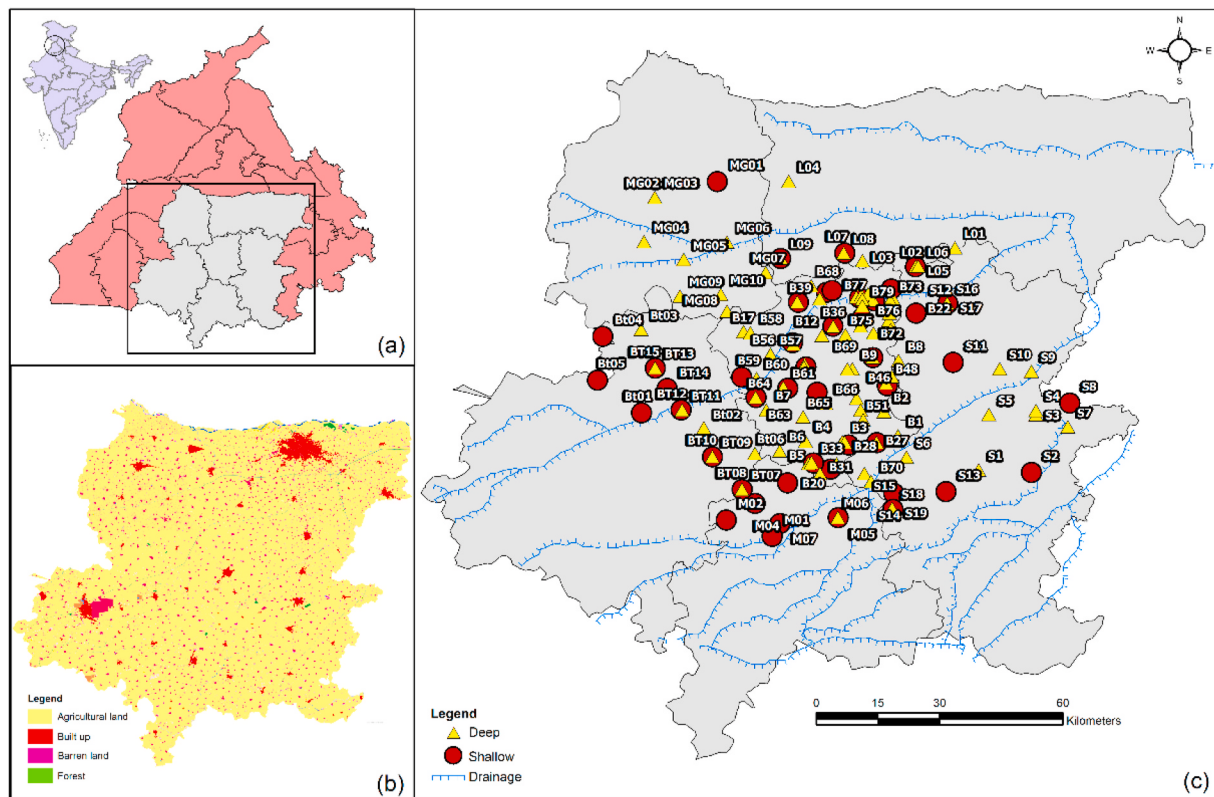


Fig. 1. (a) Location of study area (b) Land use and (c) Sampling sites.

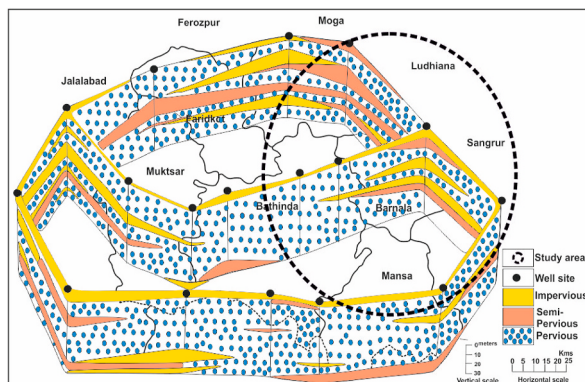


Fig. 2. Lithological Fence diagram of south-west Punjab (modified after Chopra and Krishan, 2014).

Multiple aquifer system exists up to the depth of 300 m with alternate bands of clay and medium-coarse sand. Ground water is in unconfined condition at shallow depth (up to a depth of 60 m) and under semi-confined or confined conditions in the deeper aquifer. Top aquifer is followed by clay bed of 15–35 m thickness and the deeper aquifer extends down to the depth of 300 m. The groundwater is fresh in almost whole district. North-eastern part is more elevated as compared to the south-western part thus reflects the topographic gradient and groundwater flow follows the general slope that is from north-east to south-west (CGWB, 2017a). Lithological cross-section as a fence diagram is shown in Fig. 2 (Chopra and Krishan, 2014).

2.2. Field sampling and analysis

A total of 140 groundwater samples were collected for the post-monsoon season in October–November 2019. Sampling was done on the basis of grid method and samples were collected from every 10 × 10

km grid from the Barnala district and 20 × 20 km grid from the adjoining area. However, there are some variations due to the problems faced in field such as power cuts or unavailability of the required parameters for wells. The water samples were collected from the supply wells used for drinking and/or irrigation purposes. Wells continuously used for water supply were sampled directly after allowing the wells to run for 10 min prior of collection. The samples were labelled properly along with the details of time, place and date of collection. Depth of the wells was obtained from the owner and ranges from 30 to 166 m. All the physical parameters like pH, EC, ORP and temperature were measured in the field in-situ with the help of Hanna Portable meters (HI98120 and HI98121). ORP values presented have been corrected to SHE (Standard Hydrogen Electrode) as the AgCl electrode was used for determination of ORP. The electrode probe for EC and pH were calibrated daily before the analysis. Samples were collected in the 125 ml tarson bottles after filtration through 0.45 µm syringe filter. Samples for the cations measurement and uranium analysis have been preserved by using ultrapure HNO₃. Bottles were prewashed and dried before sampling. Unfiltered water samples were also collected for stable isotope analysis in 50 ml glass bottles and were sealed tightly. The samples were freezed at 4°C until the laboratory analyses were carried out.

2.3. Laboratory analysis

Major cation-anion measurements were carried out at Panjab University laboratory, Chandigarh by using standard methods (APHA, 1995). Sodium (Na⁺) and potassium (K⁺) measurement was done with the use of flame photometer. Calcium (Ca²⁺) and the total hardness were determined by using EDTA titrimetric methods; magnesium (Mg²⁺) was then calculated theoretically. Bicarbonate (HCO₃⁻) content was measured by using HCl titrimetric method and chloride (Cl⁻) was estimated by using Mohr's argenometric titration (Trivedy et al., 1984). Sulphate (SO₄²⁻), phosphate (PO₄³⁻) and nitrate (NO₃⁻) were analyzed with the use of UV visible spectrophotometer (Spectronic 21D). Ion

charge Balance Error has been calculated and is within the acceptable limit of $\pm 10\%$.

Uranium and stable isotope measurement is done with the help of Inductively Coupled Plasma Mass Spectrometer (ICP-MS; Agilent 8900 Triple Quadrupole) and Isotope-Ratio Mass Spectrometer (IRMS; Thermo Scientific Delta V advantage) respectively at Indian Institute of Technology (IIT), Kanpur. Precision and accuracy were determined by using standard solution as replicates. The overall accuracy of the standard analyzed was $\leq 7.8\%$ for U, $\leq 0.2\%$ for $\delta^{18}\text{O}$ and $\leq 0.3\%$ for $\delta^2\text{H}$, with a precision better than $\leq 2.4\%$ for U, 0.1% for $\delta^{18}\text{O}$ and 0.5% for $\delta^2\text{H}$.

2.4. Health risk assessment

It can be done in terms of radiological toxicity as well as chemical toxicity. As uranium is a radioactive element, radiological toxicity or carcinogenic risk is measured in terms of annual effective dose (D) and Excess cancer risk (ECR) and is calculated on the basis of average life span of people in the region, their consumption of water and uranium activity concentration. It determines the probability of individual to cancer risk due to exposure to carcinogenic hazards. Being a heavy metal, it is necessary to measure the chemical toxicity or non-carcinogenic effects of uranium with target on bone and kidneys mainly. Chemical toxicity risk is evaluated in terms of Lifetime Average Daily Dose (LADD) and Hazard Quotient (HQ).

2.4.1. Annual effective dose (D)

International Commission on Radiological Protection (ICRP, 2008) has given the annual effective dose that represents the health risk to whole human body and is quantified as sum of equivalent doses in all specified tissue and organs of body. It is calculated as (USEPA, 2000):

$$D (\mu\text{Sv.y}^{-1}): A \times I \times F$$

A is the activity concentration of uranium and is taken in terms of Bq.L⁻¹ and is calculated as measured value of U in $\mu\text{g.L}^{-1}$ multiplied by conversion factor (0.025 Bq.L⁻¹). Annual ingestion i.e. I is taken as 1478.25 L.y⁻¹ at the rate of 4.05 L.day⁻¹ (HDR, 2009; Jain et al., 1995). F is taken as dose conversion factor obtained as average of dose coefficient of uranium isotopes ²³⁴U, ²³⁵U and ²³⁸U based on ICRP, 2008 and is taken as 4.63×10^{-8} Sv.Bq⁻¹.

2.4.2. Excess cancer risk (ECR)

It is estimated on the basis of USEPA standard method (USEPA, 2000a).

$$\text{ECR} = A \times r \times I$$

A is the activity concentration of uranium. r is the risk coefficient of uranium and is taken as 1.19×10^{-9} Bq.L⁻¹. Total exposure or activity intake of uranium per capita i.e. I is calculated as product of daily intake of water which is 4.05 L.day⁻¹ for an adult in India (HDR, 2009; Jain et al., 1995) and total days of exposure in average life span (365*65) of a person (WHO, 2011b).

2.4.3. Lifetime average daily dose (LADD)

It is described as quantity of chemical substance ingested in a day per kilogram of body weight. LADD is calculated in $\mu\text{g.kg}^{-1}.\text{day}^{-1}$ by using the following equation given by USEPA (2005).

$$\text{LADD} = \text{EPC} \times \text{IR} \times \text{EF} \times \text{ED} / \text{AT} \times \text{BW}$$

Exposure point concentration of uranium i.e. EPC is measured in $\mu\text{g.L}^{-1}$. IR is the ingestion rate of water and is 4.05 L.day⁻¹; Exposure frequency (EF) is taken as 350 days.y⁻¹ (USEPA, 1991); ED is the duration of exposure i.e. the life expectancy of a person and is 65 years (WHO, 2011b); AT and BW is the average time in days (365* 65) and average body weight of Indian person (51.5 kg) respectively (Dang et al.,

1994).

2.4.4. Hazard Quotient (HQ)

It is used to estimate the harm extent by using following equation (USEPA, 2005)

$$\text{HQ} = \text{LADD} / \text{RfD}$$

RfD (Reference dose) value is given by AERB, 2004 and is taken as $4.53 \mu\text{g.kg}^{-1}.\text{day}^{-1}$.

3. Results & discussion

3.1. Rock-water interaction and hydrochemical facies

Hydro chemical characterization of groundwater using piper trilinear diagram (Piper, 1944) shows that HCO_3^- is the dominant species among anions whereas $\text{Na}^+ + \text{K}^+$ is dominant among cations (Fig. 3) in both the aquifers. It indicates that $\text{Na}^+ - \text{HCO}_3^-$ and $\text{Mg}^{2+} - \text{HCO}_3^-$ are dominant groundwater types in the deeper aquifer whereas shallow groundwater has dominance of Na^+ and Mg^{2+} bicarbonate type of water with $\text{Na}^+ - \text{Cl}^- - \text{SO}_4^{2-}$ type also present in few of the samples. Mixed facies type in the shallow water results from the higher salinity due to evaporation, and mixing of surface and groundwater in the shallow aquifer. The change in groundwater type also varies with the characteristics of groundwater which changes by its interaction with aquifer sediments along with change in direction, flow path and recharge (Singh et al., 2017a).

Groundwater chemistry is also controlled by variety of hydro-geochemical processes taking place in it. To have an insight on these, gibbs diagram were prepared by plotting Total Dissolved Solids (TDS) concentration against ratio of $(\text{Na}^+ + \text{K}^+) / (\text{Na}^+ + \text{K}^+ + \text{Ca}^{2+})$ and $\text{Cl}^- / (\text{Cl}^- + \text{HCO}_3^-)$ for cations and anions respectively (Gibbs, 1970). It is seen that majority of the samples fall in the rock dominance zone resulting from interaction of groundwater and aquifer sediments in both the shallow and deeper aquifer (Fig. 4(a) and (b)). Few of the samples also lie in the evaporation dominant zone, signifying the role of evaporation particularly in case of shallow groundwater chemistry.

If silicate and carbonate weathering takes place, bicarbonate ions are usually dominant (Elango and Kannan, 2007). Na^+ vs. HCO_3^- scatter plot (Fig. 4(c)) of current study confirms the same. Rainwater and irrigated water infiltration results in dissolution of silicate & carbonate minerals present along the path and thus bicarbonate and calcium ions are released into the groundwater (Dehnavi et al., 2011). Also, calcium and magnesium ions are dominant than the bicarbonate and sulphate

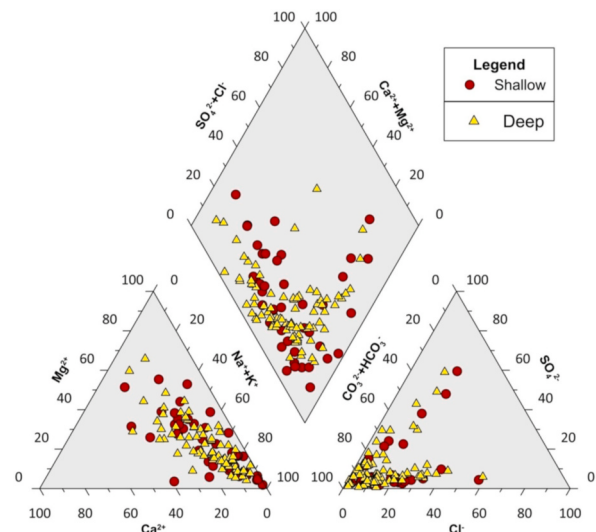


Fig. 3. Piper plots of the groundwater quality parameters.

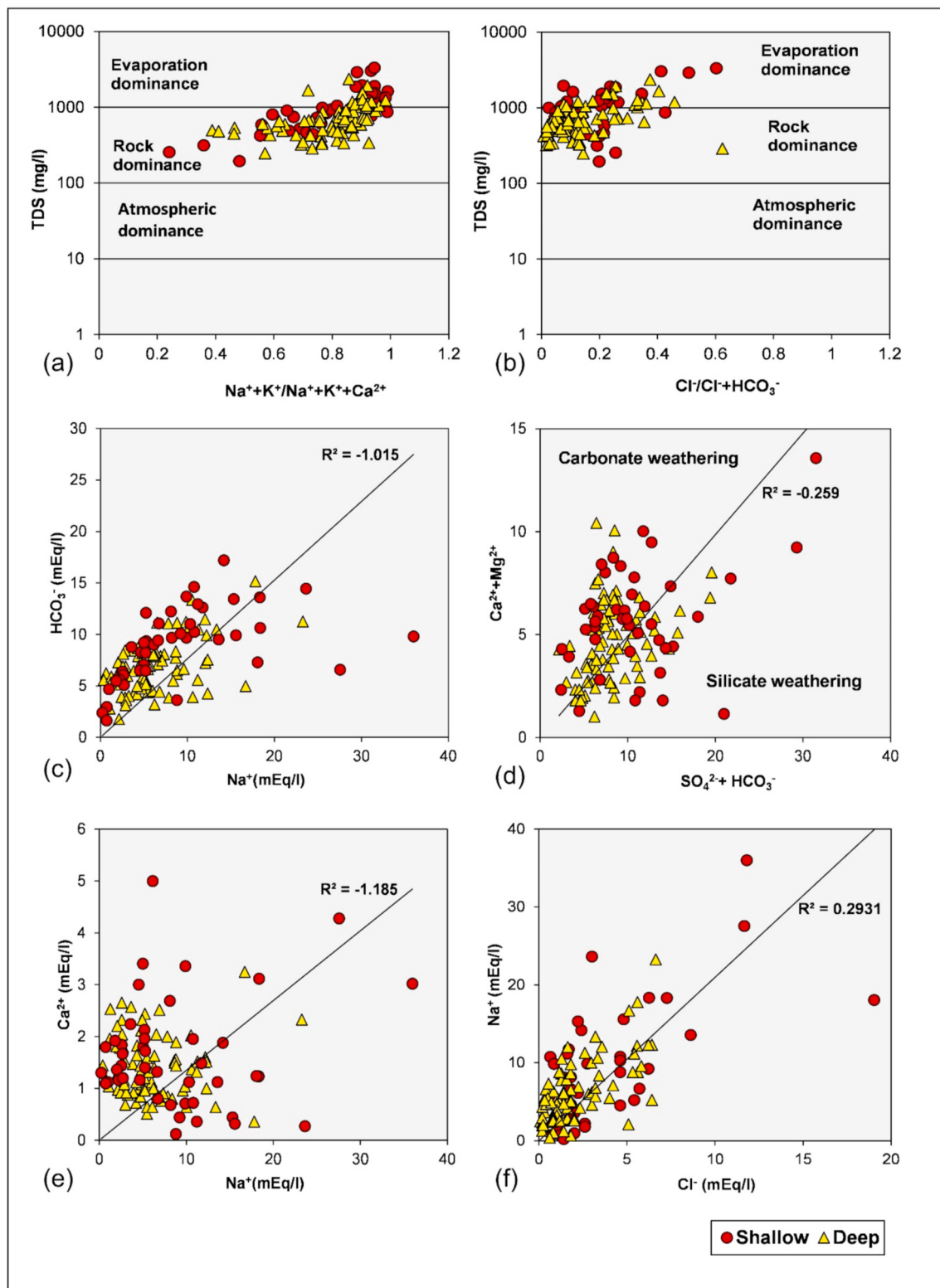


Fig. 4. (a) Scatter plot of TDS vs $(\text{Na}^+ + \text{K}^+)/(\text{Na}^+ + \text{K}^+ + \text{Ca}^{2+})$ (b) Scatter plot of TDS vs $\text{Cl}^-/(\text{Cl}^- + \text{HCO}_3^-)$ ratios (c) Plot between Na^+ vs. HCO_3^- (d) Plot between Na^+ vs Ca^{2+} (e) Scatter plot of $\text{Ca}^{2+} + \text{Mg}^{2+}$ vs $\text{SO}_4^{2-} + \text{HCO}_3^-$ (f) Plot between Na^+ vs Cl^- .

ions in the current study, with majority of samples lying above the equiline as shown in the scatter plot of $\text{Ca}^{2+} + \text{Mg}^{2+}$ versus $\text{HCO}_3^- + \text{SO}_4^{2-}$ (Fig. 4(d)). It shows that Ca^{2+} and Mg^{2+} are dominant under the influence of carbonate weathering. Samples close to the equiline shows dissolution of Calcite, dolomite or gypsum (Singh et al., 2017a,b). Few of the samples falling below the equiline show ion-exchange in the

silicate weathering zone, from albite and bicarbonate. Calcium content in the groundwater has increased from the $\text{Ca}^{2+}/\text{Na}^+$ ion exchange as shown in Na^+ vs. Ca^{2+} scatter plot (Fig. 4(e)).

Ionic species in groundwater is also affected by the factors such as evaporation. In Na^+ vs Cl^- plot, samples falling along the equiline show halite dissolution (Fig. 4(f)). In the current study most of the samples

have Na^+/Cl^- value >1 and thus show the dominance of Na^+ ions that may have resulted from the silicate weathering and/or ion exchange processes (Rajmohan and Elango, 2003). However, Na/Cl ratios are greater than what would be expected from mixing between fresh canal water and more saline groundwater. To know the process of ion exchange chloro-alkaline indices are employed (Schoeller, 1977) and negative value of indices indicate that indirect or reverse ion exchange of Ca^{2+} & Mg^{2+} in groundwater takes place with Na^+ and K^+ of aquifer material. The role of reverse-ion exchange processes and carbonate weathering is also observed in the adjoining areas (Kumar et al., 2011; Rishi et al., 2017).

3.2. Hydrochemistry and distribution of ions

Descriptive statistics of the groundwater quality parameters were determined for both the shallow and deep water and statistical summary is given in Table 1 along with the WHO recommended values. Based on the mean concentration of anions also, HCO_3^- is found to be the most dominant anion followed by $\text{Cl}^- > \text{SO}_4^{2-} > \text{NO}_3^-$ and among cations Na^+ is found as the dominant cation followed by $\text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+$ in both the aquifers. The pH value ranges from 6.95 to 8.27 thus indicating the alkaline nature of groundwater. Electrical conductivity (EC) of the groundwater ranges from 290 to 4970 $\mu\text{S}\cdot\text{cm}^{-1}$ and is higher in the shallow groundwater. Higher oxidation-reduction potential (ORP) values are found in the shallow aquifers owing its access to oxygen. In the deeper aquifer, lower ORP values are found in less than 5% samples, representing anoxic conditions, however, in deeper aquifer also oxic conditions are prevalent (Figs. 5–7). The range described is concordant with the value observed by Kumar et al. (2021) in Bathinda, Barnala and Ludhiana; Sharma et al. (2021) in Mansa district, Ahada and Suthar

Table 1
Descriptive statistics of groundwater quality parameters and WHO limit.

	Shallow Aquifer					WHO (2011)
	Mean	Median	SD	Minimum	Maximum	
pH	7.46	7.5	0.27	7.05	8.24	6.5–8.5
EC	1649.73	1375	568.68	290	4970	1500
ORP	333.52	346.5	5.91	224	396	—
Na^+	204.50	153	94.21	4.90	827.12	200
K^+	9.31	6.5	2.64	2.70	42.41	30
Ca^{2+}	32.97	27.66	11.53	2.40	100.20	200
Mg^{2+}	52.31	49.21	20.29	9.26	172.05	150
HCO_3^-	540.98	566	149.93	100	1050	300
PO_4^{3-}	0.14	0.04	0.03	0.01	1.03	0.1
NO_3^-	58.07	23.5	68.97	8.80	259.44	50
SO_4^{2-}	99.39	29.11	98.82	5.20	1196.38	250
Cl^-	124.70	71	64.01	22.39	674.50	250
U	85.43	63.2	23.35	4.26	318.03	15
	Deep Aquifer					WHO, 2011
	Mean	Median	SD	Minimum	Maximum	
pH	7.51	7.49	0.21	6.95	8.27	6.5–8.5
EC	1105.90	980	1078.22	370	3520	1500
ORP	298.51	310	7.03	92	413	—
Na^+	134.93	114.6	173.95	8.70	534.81	200
K^+	5.74	5.75	8.26	0.10	21.94	30
Ca^{2+}	28.34	26.96	21.32	7.21	65.06	200
Mg^{2+}	41.22	35.92	31.55	6.22	97.46	150
HCO_3^-	427.73	420	216.27	108.73	925	300
PO_4^{3-}	0.23	0.07	0.03	0.01	1.11	0.1
NO_3^-	25.92	19.15	51.20	0.08	112.76	50
SO_4^{2-}	59.70	30.9	233.17	3.05	701.48	250
Cl^-	73.35	52.62	127.98	2.52	234.92	250
U	43.50	36.68	66.40	8.11	129.73	15

Note: All the units are in $\text{mg}\cdot\text{L}^{-1}$ except pH, EC ($\mu\text{S}\cdot\text{cm}^{-1}$), ORP (mV) and U ($\mu\text{g}\cdot\text{L}^{-1}$). WHO,

World Health Organization; SD, Standard Deviation; EC, electrical conductivity; ORP, oxidation-reduction potential.

(2018) in south-west Punjab.

Spatial distribution maps and the vertical graphs (Figs. 5–7) shows that concentration of ions is higher in the southern part and shallow aquifer of the region. Climate plays an important role in the increasing concentration towards southern portion. Rainfall decreases in the south-west direction and aridity increases resulting in higher evaporation (CGWB, 2017b; Paul et al., 2015; Sidhu et al., 2021). This leads to the high conductivity and higher ion concentration in the south. Higher concentration observed at shallow depth may have resulted due to the soil mineralization with rapid recharge of water through shallow aquifers along with anthropogenic input whereas source of major ions in the deeper aquifers is lithogenic, resulting from dissolution of rocks & minerals of aquifer sediments. Dissolved ion concentration in the groundwater depends on the aquifer matrix and their solubility (Sarin et al., 1989). Addition of nutrients from mineral fertilizers in farmland areas also increases the EC of shallow groundwater.

3.3. Distribution of uranium

Uranium is among the major contaminants that pose severe health risk to population. Uranium concentration was found to be present in 76% of the water samples higher than the detection limit of World Health Organization (WHO, 2011a), $30\text{ }\mu\text{g}\cdot\text{L}^{-1}$. 34% of samples have uranium concentration greater than or equal to the drinking water guideline provided by Atomic Energy Regulatory Board (AERB) of $60\text{ }\mu\text{g}\cdot\text{L}^{-1}$. Uranium concentration ranges from 4.26 to $318.03\text{ }\mu\text{g}\cdot\text{L}^{-1}$ and is higher at shallow depth. The range described is comparable to the uranium concentration reported by previous researchers in Punjab (Bajwa et al., 2015; Coyte et al., 2018; Pant et al., 2017; Rishi et al., 2017; Singh et al., 1995; Singh and Kishore, 2010; Saini et al., 2016; Sharma et al., 2017) and Barnala (Kumar et al., 2021; Virk, 2019). In present study, it is seen that uranium distribution is not homogenous over the entire area (Figs. 5 and 6) and among all the samples exceeding recommended values, highest concentration is observed in the shallow water of Bathinda district followed by Mansa, Sangrur, Barnala and Ludhiana respectively. In case of deeper aquifer, highest contamination is seen in case of Moga district followed by Sangrur, Bathinda, Ludhiana and Barnala respectively. Uranium levels are reported in all the districts in Table 2.

An increasing trend of contamination is observed moving from north-east to south-west and hotspots are prominent in the southern region. Increase in uranium concentration towards the south-west can be due to the following reasons. In the study region, groundwater flow is towards the low-lying terrain in the south-west direction (CGWB, 2017b; Krishan et al., 2021) and has resulted in higher uranium concentration due to the longer time gap available for water-rock interactions. Increase of uranium concentration along the groundwater flow direction is also observed in the Al-Batin alluvial fan aquifer of south Iraq by Alkinani et al. (2016). Climate also plays an important role in the higher uranium levels; south-west region is comparatively dry due to less rainfall and higher evaporation (CGWB, 2017b; Paul et al., 2015; Sidhu et al., 2021). Higher uranium concentration in the dry climate is also observed by other researchers (Ayotte et al., 2011). Groundwater logging and nearly disrupted cycle of groundwater recharge in the south-west region (PC, 2013) is responsible for increased concentration of uranium in groundwater. As irrigation and drinking water supply is mainly based on canals in this part and there is minimal use of groundwater due to the increased salinity (Krishan et al., 2021). Also the water recharge from unlined canal network and flood irrigation increases the water level in the south-west direction (Baweja et al., 2017) and results in increased redox potential of water with more oxygen content and thus increases the uranium concentration in groundwater because of its higher solubility and mobility in oxidized hexavalent (U(VI)) form.

Uranium concentration levels are also strongly correlated with the depth of wells and concentration decreases with increase in depth. Though precise pattern varies from region to region, usually wells

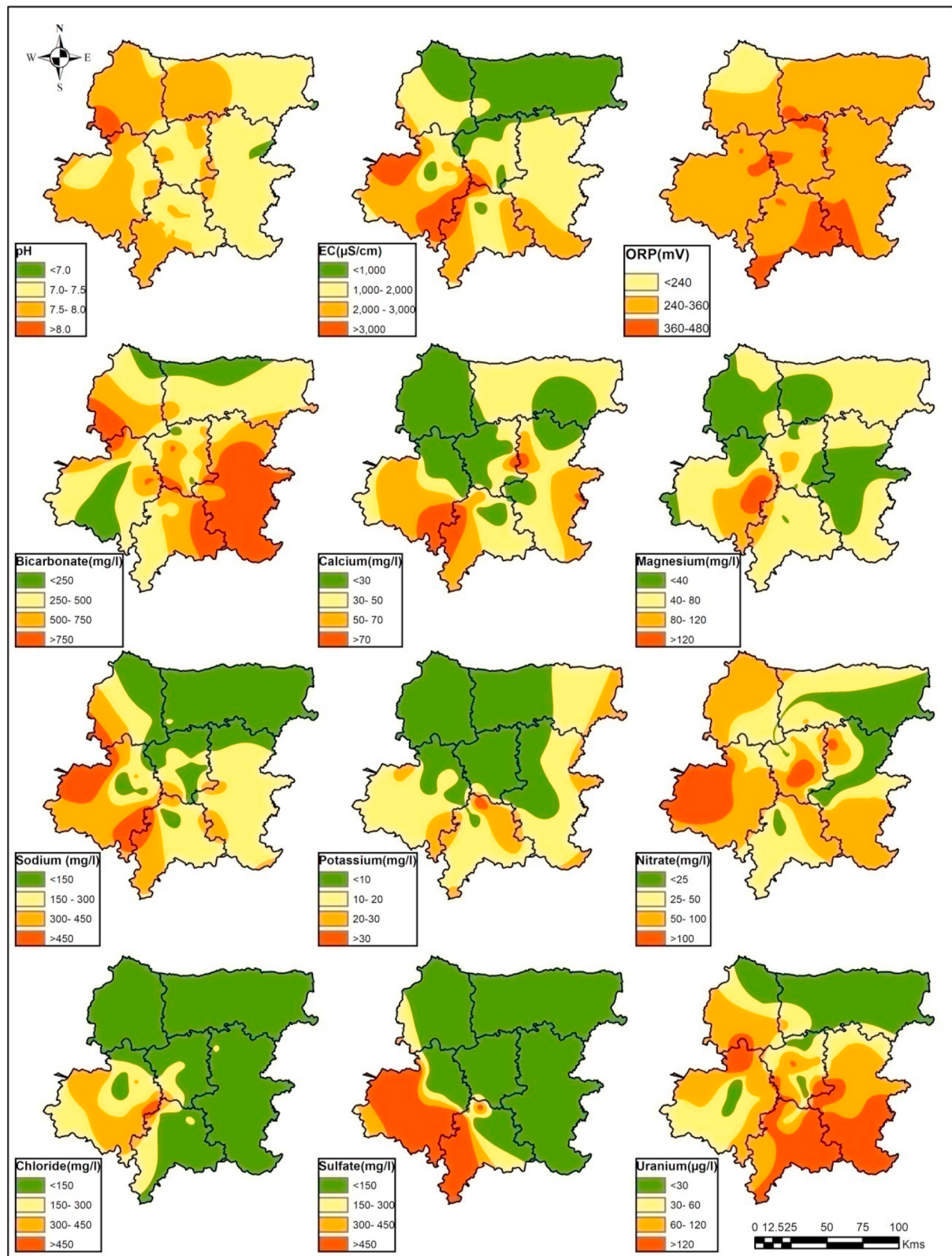


Fig. 5. Spatial distribution of pH, EC, ORP, U and major ions in the shallow groundwater of study region.

deeper than 60 m are less contaminated and higher uranium concentration (>AERB permissible limit) is confined to the shallow aquifer only in the top 60 m. Similar results have also been reported by researchers in south-west Punjab (Saini et al., 2016; Sharma et al., 2017). Anthropogenic inputs such as irrigation return flow (Sahoo et al., 2021), input

from fertilizers (nitrate & phosphate) (Alrakabi et al., 2012; Tripathi et al., 2013) in the shallow water has resulted in higher concentration of major ions such as bicarbonate, electrical conductivity (Figs. 5 and 7) and thus results in increased mobilization of uranium in a way described below.

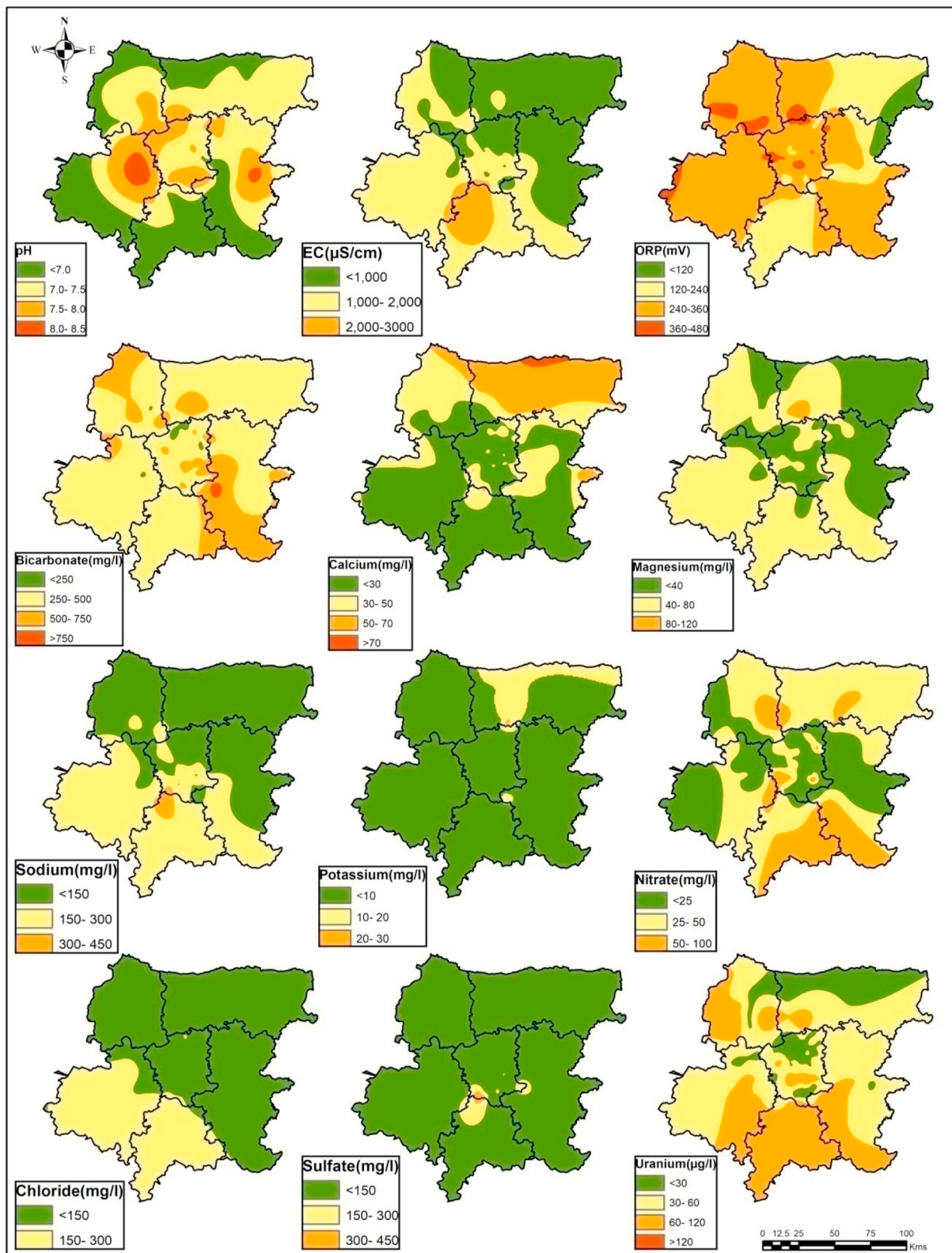


Fig. 6. Spatial distribution of pH, EC, ORP, U and major ions in the deep groundwater of study region.

3.4. Geochemical controls of uranium in groundwater

Uranium in the groundwater may be present from the natural processes or anthropogenic inputs that results in its increased mobilization in the sub-surface (Coyle et al., 2018; Smedley et al., 2006). In the present study uranium in groundwater is predicted from the basic

geochemistry parameters of water. The chemical parameters used in the current study are major ion composition, pH, EC and ORP. Knowledge of groundwater chemistry is essential to understand its influence on the release of uranium from sediments. Data being non-parametric in nature, spearman rank correlation (Table 3) is employed to define the relationship between uranium and other physico-chemical parameters.

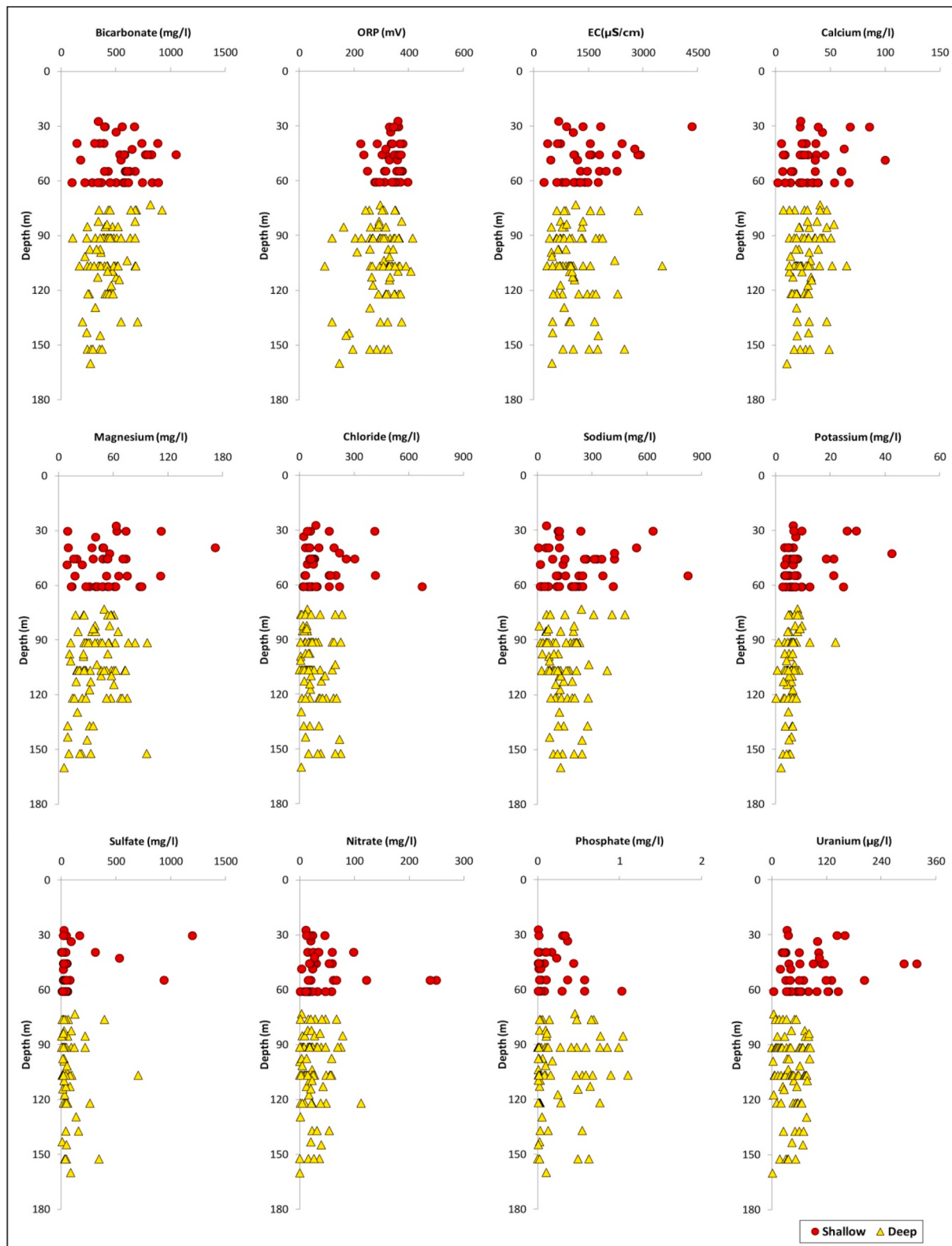


Fig. 7. Vertical distribution of groundwater quality parameters in the shallow and deeper groundwater.

Usually, pH is paramount in governing the dissolution of uranium in the groundwater and affects uranium speciation (Echevarria et al., 2001). Overall pH in the current study is alkaline (>7) and alkaline pH, increases the uranium desorption leading to its dissolution (Wu et al., 2014). Uranium release from soil matrix also depends on the oxidation and reduction reactions taking place (oxidation-reduction potential).

Stable uranium minerals form in the reducing environment (low ORP), causing uranium to precipitate out of the groundwater whereas oxygenated groundwater (high ORP) helps in its dissolution and transportation (Alam and Cheng, 2014; Szecsody et al., 1998). High uranium concentration in oxidizing groundwater of both shallow and deeper aquifer is evident from Figs. 7 and 8. Important role of pH and ORP has

Table 2

District-wise summary of uranium levels in shallow and deep groundwater of study region along with percentage of samples higher than the prescribed limit.

District/Aquifer (no. of samples)		Median	Minimum	Maximum	>WHO limit	>AERB limit
Sangrur (19)	Shallow (7)	91.51	54.61	318.03	100%	86%
	Deep (12)	48.98	26.51	107.86	92%	17%
Barnala (68)	Shallow (19)	41.47	18.78	290.55	84%	37%
	Deep (49)	32.78	8.1	129.72	58%	12%
Bathinda (15)	Shallow (8)	109.11	4.26	204.05	88%	88%
	Deep (7)	43.69	BDL	71.14	58%	14%
Ludhiana (10)	Shallow (3)	36.72	32.28	61.87	100%	33%
	Deep (7)	35.46	25.67	84.88	58%	14%
Mansa (7)	Shallow (6)	104.3	33.89	160.03	100%	84%
	Deep (1)	NA	NA	NA	NA	NA
Moga (10)	Shallow (1)	NA	NA	NA	NA	NA
	Deep (9)	49.41	27.01	102.75	88%	33%

Note: Units are in $\mu\text{g.L}^{-1}$. NA, Not Applicable; BDL, Below Detection Limit; WHO, World Health Organization; AERB, Atomic Energy Regulatory Board of India.**Table 3**

Spearman rank correlation of uranium and physico-chemical parameters in the shallow and deep aquifer of study region.

	Shallow Aquifer												
	pH	EC	ORP	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	HCO ₃ ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	Cl ⁻	U
pH	1.00												
EC	-0.47	1.00											
ORP	-0.13	0.10	1.00										
Na ⁺	-0.34	0.91 ^a	0.01	1.00									
K ⁺	-0.59 ^a	0.41 ^a	0.16	0.35 ^b	1.00								
Ca ²⁺	-0.56 ^a	-0.05	-0.02	-0.13	0.55 ^a	1.00							
Mg ²⁺	-0.31	0.28	0.17	0.01	0.42 ^a	0.16	1.00						
HCO ₃ ⁻	-0.50 ^b	0.69 ^a	0.16	0.75 ^a	0.36 ^b	-0.06	-0.01	1.00					
NO ₃ ⁻	-0.10	0.43 ^a	-0.04	0.35 ^b	0.01	0.04	0.15	0.24	1.00				
SO ₄ ²⁻	-0.27	0.68 ^a	0.14	0.75 ^a	0.32 ^b	-0.05	0.15	-0.52 ^a	0.22	1.00			
PO ₄ ³⁻	-0.61 ^a	0.16	0.17	0.29	0.135	0.27	-0.14	0.27	0.08	0.35 ^b	1.00		
Cl ⁻	-0.07	0.70 ^a	0.06	0.57 ^a	0.29 ^b	-0.10	0.47 ^a	0.31 ^b	0.38 ^b	0.49 ^a	-0.26	1.00	
U	-0.51 ^b	0.80 ^a	0.14	0.84 ^a	0.41 ^a	-0.14	0.07	0.77 ^a	0.14	0.74 ^a	0.19	0.52 ^a	1.00

	Deep Aquifer												
	pH	EC	ORP	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	HCO ₃ ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻	Cl ⁻	U
pH	1.00												
EC	-0.42 ^a	-0.16											
ORP	-0.16	0.07	1.00										
Na ⁺	-0.07	0.82 ^a	-0.02	1.00									
K ⁺	-0.47 ^a	0.24 ^b	-0.14	0.07	1.00								
Ca ²⁺	-0.61 ^a	0.19	-0.22 ^b	-0.13	0.56 ^a	1.00							
Mg ²⁺	-0.54 ^a	0.43 ^a	0.33 ^a	-0.01	0.37 ^a	0.34 ^a	1.00						
HCO ₃ ⁻	-0.59 ^b	0.58 ^a	0.14	0.41 ^a	0.49 ^a	0.34 ^a	0.59 ^a	1.00					
NO ₃ ⁻	-0.26	0.30 ^a	-0.12	0.19	0.05	0.05	0.32 ^a	0.31 ^a	1.00				
SO ₄ ²⁻	-0.15	0.54 ^a	-0.19	0.71 ^a	0.07 ^b	-0.01	-0.23 ^b	0.12	-0.03	1.00			
PO ₄ ³⁻	0.04	-0.06	-0.52 ^a	-0.02	0.23 ^b	0.39 ^a	-0.14	0.13	-0.05	0.26 ^b	1.00		
Cl ⁻	-0.27	0.73 ^a	0.33 ^a	0.60 ^a	-0.05	-0.10	0.37 ^a	0.17	0.30 ^a	0.21 ^b	-0.48 ^a	1.00	
U	-0.51 ^a	0.49 ^a	0.17	0.41 ^a	0.45 ^a	-0.21	0.48 ^a	0.78 ^a	0.21	0.10	0.02	0.18	1.00

^a Correlation significant at level 0.01.^b Correlation significant at 0.05 level; EC, Electrical Conductivity, ORP, Oxidation-reduction potential.

been considered by Kumar et al. (2011) and Hundal (2011) in adjoining districts. In the current study it is observed that pH and ORP provides favourable conditions for the release of uranium but are not the controlling factors of uranium mobilization because of oxic and alkaline nature of groundwater in the entire region.

U is seen to have positive correlation with electrical conductivity, bicarbonate, sodium and potassium in both the aquifers (Fig. 8, Table 3). It is generally believed that higher the conductance, higher will be the radioactivity of water. Uranium sorption to the sediment decreases with increase in ionic strength because of higher competition among ions in the groundwater. When ionic strength of oxidized solution increases, uranyl ion displacement takes place from the soil exchange sites by other ions such as Ca²⁺, Mg²⁺ and K⁺ and results in its mobilization in high ionic strength solution (Kumar et al., 2014; Krupa et al., 1998; Li et al., 2016). Very strong correlation (p = 0.80) of uranium is seen with EC in shallow aquifer and is moderate (p = 0.49) in the deeper aquifer. This is consistent with the higher EC value of shallow aquifer due to input from

irrigation recharge, infiltration of rainwater and nutrient addition by fertilisers. Thus greater mobilization is seen in the shallow aquifer and EC has strong effect on uranium mobilization in semi-arid regions. U is strongly correlated with Na⁺ (p = 0.84) in the shallow aquifer and is moderately correlated in deeper aquifer (p = 0.41), however, the values are comparable in case of K⁺ (p = 0.41 & 0.45) in both the aquifers. This shows that liberation of these ions also has effect on uranium mobilization. Leaching and migration of U in alkali matrix is also reported by the Dressen et al. (1982). Strong positive correlation of uranium has also been observed with sulphate (p = 0.74) in the shallow aquifer as higher sulphate value is observed in the shallow aquifer and it can act as competing ion for U species (Wu et al., 2018).

In U affected areas, bicarbonate is a major factor controlling uranium mobilization (Langmuir, 1978). In the study region, concordant changes are observed in the uranium-bicarbonate pattern (Figs. 7 and 8) and very strong positive correlation of uranium & bicarbonate has been observed in both the aquifers (p = 0.77 & 0.78). Intensive agriculture in the region

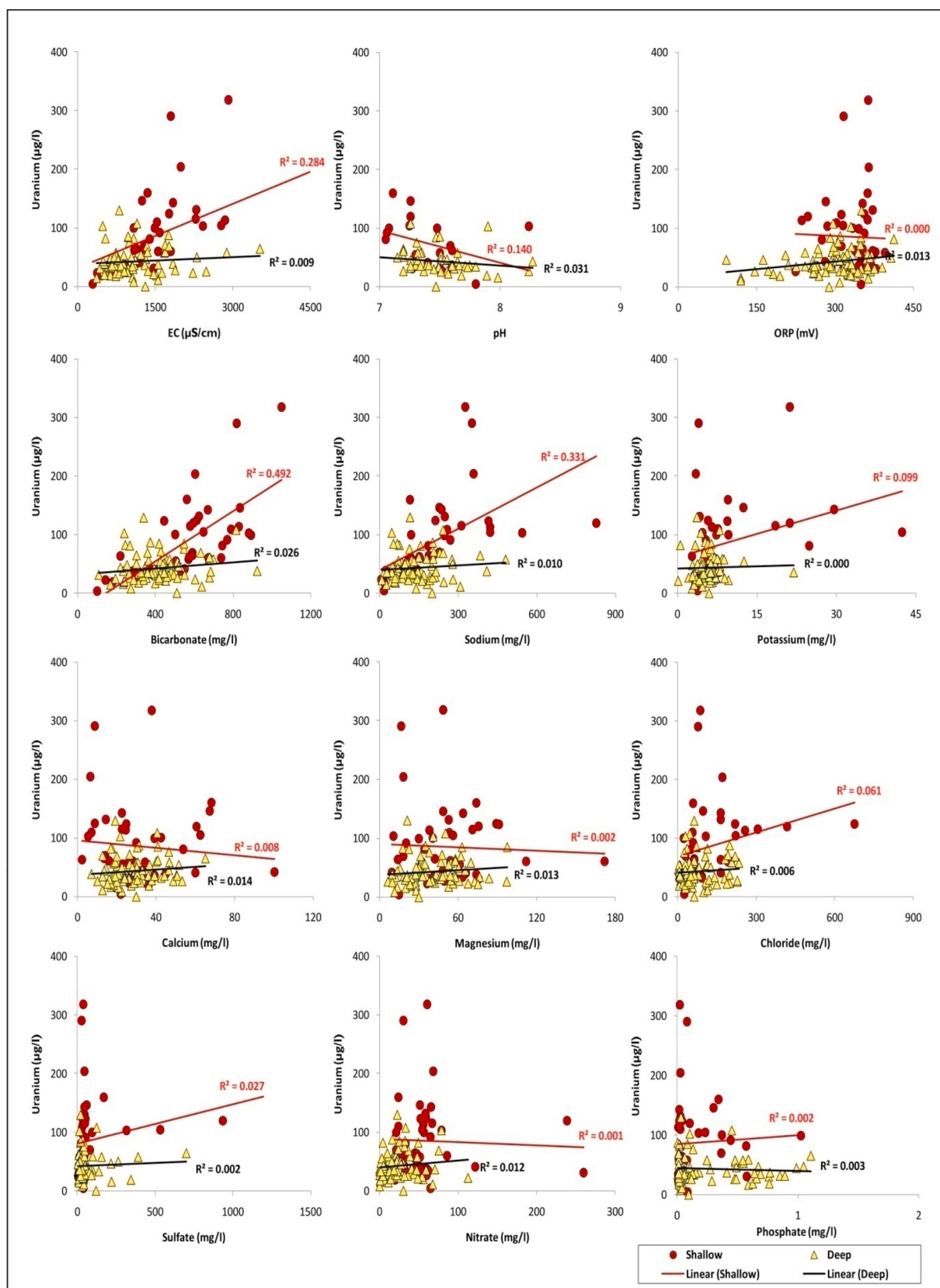


Fig. 8. Association of uranium with basic groundwater parameters.

have resulted in the high bicarbonate present in the groundwater due to plant root respiration that increases the amount of $\text{CO}_2(\text{g})$ in the soil resulting in formation of bicarbonate ions (Rishi et al., 2017). Bicarbonate ions form soluble and stable complexes with uranyl ions that lead to leaching of uranium from the sediments and its migration to long distances (Langmuir, 1997). Uranium mobilization due to the formation of carbonate complexes is known worldwide from the speciation results for similar groundwater environments (Krestou and Panias, 2004; Kumar et al., 2014; Nair et al., 2014; Smedley et al., 2006; Wu et al., 2014). Use of bicarbonate rich groundwater for irrigation also results in higher mobilization of uranium to groundwater (Burow et al., 2017; Jurgens et al., 2008, 2010). Carbonic water percolation through calcareous soils increases its efficiency of leaching uranium from soils. Uranium mobilization in the oxidized groundwater of alkaline pH due to higher levels of bicarbonate has also been reported by researchers from adjoining regions (Pant et al., 2017, 2019; Rishi et al., 2017; Sharma et al., 2017).

3.5. Environmental stable isotopes

Source and mechanism of groundwater recharge was identified by employing stable isotopes and were related with the uranium contamination. Stable isotope content is plotted as $\delta^2\text{H}$ and $\delta^{18}\text{O}$ and compared with the reference Global Meteoric Water Line (GMWL) (Craig, 1961). Isotope show wide variation with $\delta^2\text{H}$ content lying in the range of -71.12 to -22.77 with an average of -44.31 . $\delta^{18}\text{O}$ content vary from -10.71 to -0.34 with average of -5.37 . All the groundwater samples fall below the GMWL and IMWL indicating the effect of evaporation

(Fig. 9(a)). The slope of best fit line indicates that the aquifers get water contribution either from irrigation return flow or from evaporated surface water body. Slope of value less than 8 shows the evaporation effect in groundwater samples (Clark and Fritz, 1997). Precipitation as major source of groundwater recharge and contribution of irrigation return flow in the shallow aquifer is reported by various workers (Keesari et al., 2017; Rao et al., 2017; Sharma et al., 2017). Contribution by canal seepage of depleted isotopic composition (Rai et al., 2014) is also reported by Tripathi et al. (2016). However, less effect of evaporation is seen in the deeper aquifer and is not a case in the current study. There was no significant difference observed in the isotope data of shallow and deeper aquifers, showing the effect of evaporation at both the levels. Thus it shows that some interconnection exists between shallow and deep aquifer, making it susceptible to the anthropogenic impacts.

Recharge and discharge considerably affects the uranium concentration in groundwater (Brindha and Elango, 2013). Water recharge from rainfall increases the water level thus also increases the uranium concentration in groundwater. Recharging fresh water interacts with the uranium enriched soil of the region and results in its dissolution. However, uranium concentration in groundwater starts to reduce due to dilution by continuing recharge of comparatively fresh water. $\delta^{18}\text{O}$ versus U plot in Fig. 9(b) shows that contaminated and uncontaminated groundwater cannot be differentiated on the basis of isotope data.

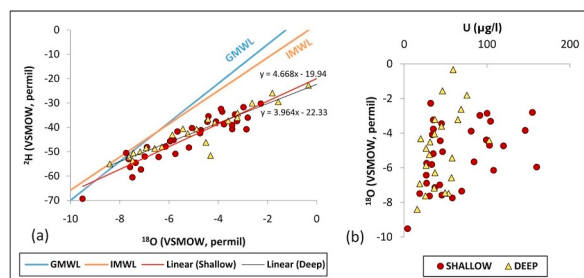


Fig. 9. (a) Stable isotope variation (b) Variation of oxygen content with uranium concentration in both shallow and deeper aquifer.

3.6. Proposed hypothesis for the source of uranium in groundwater

A lot of studies have been carried out regarding the source of uranium in the most affected Punjab state, however, facts are still not clear and requires more work. Research that has been done till date indicate towards geogenic origin of uranium (Patnaik et al., 2016; Singh et al., 2009; Sharma et al., 2018; Tripathi et al., 2013). Current study provides a schematic representation of uranium mobilization from the Siwaliks lying on the north-east of Punjab (Fig. 10(a)). Siwalik sediments are the fluvial deposits along the foothills of Himalayas and are formed of the detritus from sedimentary, igneous, and metamorphic rocks (Krynine, 1937). These granitic bodies have supplied the sediment to form alluvial plains of Punjab lying at foothills of Siwaliks. Primary and secondary uranium minerals are reported from the tertiary and older sedimentary rocks of Siwaliks and are restricted to the uranium mineralized zones (Kaul et al., 1993; Phadke et al., 1985). Siwaliks contributes to the most of groundwater recharge in the region and meteoric origin groundwater is supposed to supply uranium due to the rock-water interactions (Dhanaraju et al., 1985; Welch and Lico, 1998). Uranium mineralization zones of Siwaliks follow the general trend of drainage channels (Rojas, 1989) and oxygenated groundwater percolating through these zones oxidizes and mobilizes uranium from the sediments. When uranium (U (VI)) enriched groundwater moves through former river channels from basin edge to basin centre (Fig. 10(a)), uranium reduction takes place from the water flowing under anaerobic conditions of past time in the favourable environment. Environment with deficiency of oxygen containing less permeable material and reducing agents such as carbonaceous material, hydrogen sulphide and pyrite is considered as the most favourable geologic environment for the uranium ore deposition (Rojas, 1989).

In present day, changing climate and land use have greater role on groundwater uranium concentration. Uranium concentration reaching up to the 10 mg.kg^{-1} is observed in the aquifer sediments of region (Bala et al., 2021). Prevailing oxidizing and alkaline water results in oxidative mobilization of U(IV) from the sediments by interaction with uranium phases (present as secondary minerals or in the form of adsorption). Mobilization is enhanced in the saline water with high bicarbonate resulting from carbonate weathering, high Na^+ and K^+ concentration resulting from the silicate weathering and reverse ion exchange processes from the aquifer sediments. Elevated uranium levels in the shallow groundwater are due to the influence of various anthropogenic activities and addition of water from multiple sources such as canal water, rain water and irrigation return flow. Continuous addition of agrochemicals and recharge of highly ionic irrigation water affects the shallow groundwater (Fig. 10(b)) and results in mobilization of uranium.

3.7. Health risk indices

Health risk assessment is calculated to determine the health effects to the people of malwa region due to ingestion of high uranium water. Human beings are continuously exposed to the uranium via food, soil, air and drinking water. In the study region, drinking water requirements are mainly fulfilled by the use of groundwater. Thus presence of uranium in the water has raised a serious health concern. Punjab has highest number of cancer patients and uranium is considered to be one of the factors causing cancer to the people of the region because of its known carcinogenic effect (Saini et al., 2016).

Cancer risk value in the region varies from 0.12×10^{-4} to 9.09×10^{-4} and AERB has recommended maximum permissible limit of 1.67×10^{-4} for excess cancer risk. Thus in present study 33% of the samples has cancer risk above the permissible limit. Annual effective dose level or Individual Dose Criterion (IDC) due to the water consumption in individual is recommended as $100 \mu\text{Sv.y}^{-1}$ (WHO, 2011b). Current dose level is 7.29–544.17 crossing the safe limit.

Lifetime daily dose due to the intake of uranium varies from 0.33 to

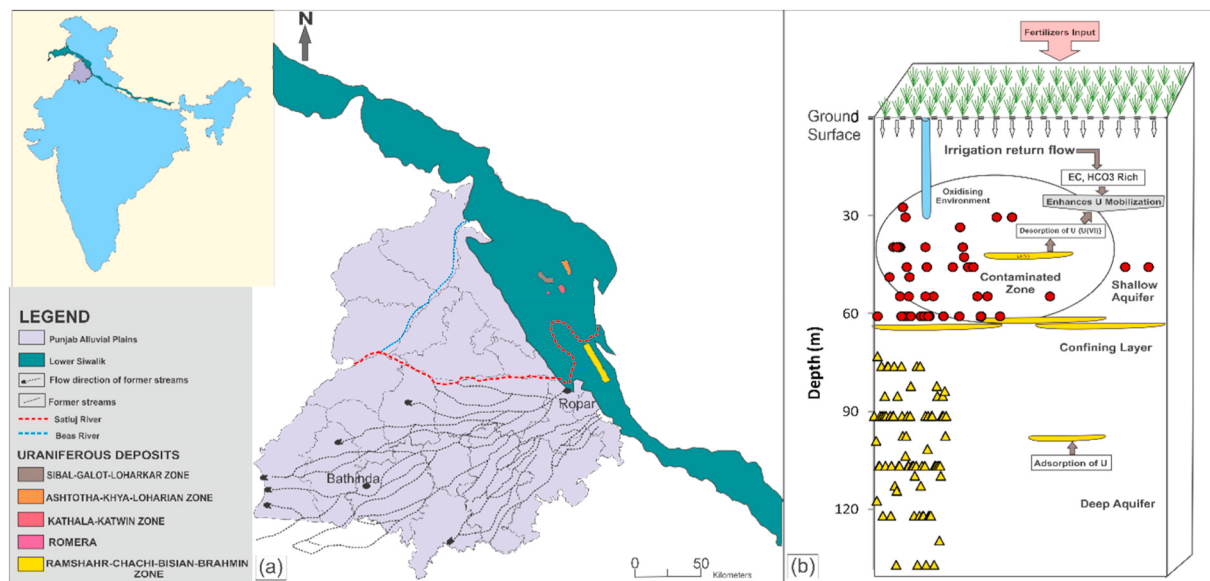


Fig. 10. (a) Uranium Zones in Siwalik (in the Beas-SatluJ valley by AMD, IAEA) and former courses of SatluJ river in NW India (after Kar, 1999) (b) Schematic diagram of uranium mobilization under present-day conditions.

25.01; contrast in the dose is mainly because of the heterogeneous distribution of uranium in the region. Around 40% of samples have LADD higher than the recommended levels of $4.53 \mu\text{g.kg}^{-1}.\text{day}^{-1}$ (AERB, 2004). Hazard quotient value also varies from 0.074 to 5.521 and value greater than 1 is not acceptable as far as the chemical toxicity is concerned. Thus groundwater in the region is not safe for drinking purposes with 33% of samples exceeding the safe limit.

4. Conclusion

This study investigates the occurrence of uranium in the alluvial aquifers of semi-arid Punjab and defines factors controlling uranium enrichment. The key findings of this study are summarized below:

- Study shows that uranium distribution is not homogenous throughout the region, however, spatial distribution graphs reveal that concentration increases towards south-west direction. Factors such as regional groundwater flow, high groundwater table due to water logging, extensive use of canal water and higher evaporation in the semi-arid south-west portion controls the enhanced uranium levels in groundwater.
- Hydrochemical analysis shows that major cations & anions concentration is higher at shallow depth along with the high uranium prominent in the shallow zones. EC, HCO_3^- , Na^+ and K^+ are identified as the controlling factors of uranium mobilization in both the aquifers. However, correlation of SO_4^{2-} and Cl^- is seen only in the shallow aquifer and gives indication towards anthropogenic enrichment. Anthropogenic inputs from fertilizers and irrigation plays vital role in the increased uranium concentration of shallow aquifer and cannot be ignored.
- Carbonate weathering is the dominant hydro-geochemical process controlling groundwater chemistry and strong correlation of uranium with electrical conductivity and alkalinity imply that weathering of carbonates and desorption of uranium from mineral surface is major cause of uranium mobilization in the aquifer system.
- Environmental stable isotopes show the evaporation signature in both the aquifers indicating the contribution of irrigation or evaporated water to the aquifer system. It also shows that a connection exists between the shallow and deeper aquifer and warns against the possible contamination of deeper aquifer in the future.

- Hazard index value exceeds the permissible limit in 33% of the samples and is a potential threat for people residing in the region by posing significant cancer risk.
- As the spatial variability of the uranium contaminated wells is very high, there is urgent need to identify the unsafe wells with large scale testing to reduce health risk. Shallow groundwater of the region is not suitable for domestic or irrigation use due to uranium contamination, however, deep wells are perhaps better option with the effective governance and by preventing mixing of shallow water to the deeper aquifer.
- Though groundwater chemistry helps in basic understanding of uranium behaviour in groundwater, much detailed aquifer sediments study is required to clearly define the source of uranium in study area and is a part of our future dynamics. Hypothesis provided regarding the source of uranium in groundwater will be useful for the future studies in this direction and can be extrapolated to the other semi-arid regions with the similar groundwater conditions.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements:

The authors are thankful to the Science and Engineering Research Board (SERB), Department of Science & Technology (DST), Government of India, New Delhi for the funding provided for current research. Authors are also grateful to the Chairperson, Department of Geology, Panjab University, Chandigarh for providing the infrastructural facilities. We are also thankful to the anonymous reviewers and editors for their valuable comments to the manuscript.

References

- Aerb, D., 2004. Drinking Water Specifications in India. Atomic Energy Regulatory Board, Mumbai.
- Ahada, C.P., Suthar, S., 2018. Assessing groundwater hydrochemistry of Malwa Punjab, India. Arabian J. Geosci. 11 (2), 1–15. <https://doi.org/10.1007/s12517-017-3355-8>.
- Alam, M.S., Cheng, T., 2014. Uranium release from sediment to groundwater: influence of water chemistry and insights into release mechanisms. J. Contam. Hydrol. 164, 72–87. <https://doi.org/10.1016/j.jconhyd.2014.06.001>.

- Alkanini, M., Kanoua, W., Merkel, B., 2016. Uranium in groundwater of the Al-Batin Alluvial Fan aquifer, south Iraq. *Environ. Earth Sci.* 75 (10), 869. <https://doi.org/10.1007/s12665-016-5685-3>.
- Alrakabi, M., Singh, G., Bhalla, A., Kumar, S., Kumar, S., Srivastava, A., Rai, B., Singh, N., Shahi, J., Mehta, D., 2012. Study of uranium contamination of ground water in Punjab state in India using X-ray fluorescence technique. *J. Radioanal. Nucl. Chem.* 294 (2), 221–227. <https://doi.org/10.1007/s10967-011-1585-x>.
- APHA, 1995. *Standard Methods for the Examination of Water and Wastewater*, nineteenth ed. American Public Association, Washington, DC.
- Ayotte, J.D., Gronberg, J.M., Apodaca, L.E., 2011. Trace Elements and Radon in Groundwater across the United States, 1992–2003. US Department of the Interior, US Geological Survey, p. 115. <https://doi.org/10.3133/sir20115059>.
- Babu, M.S., Somashekar, R.K., Kumar, S.A., Shivanna, K., Krishnamurthy, V., Eappen, K. P., 2008. Concentration of uranium levels in groundwater. *Int. J. Environ. Sci. Technol.* 5 (2), 263–266.
- Bajwa, B.S., Kumar, S., Singh, S., Sahoo, S.K., Tripathi, R.M., 2015. Uranium and other heavy toxic elements distribution in the drinking water samples of SW-Punjab, India. *J. Radiat. Res. Appl. Sci.* 10 (1), 13–19. <https://doi.org/10.1016/j.jrras.2015.01.002>.
- Bala, R., Karanveer Das, D., 2021. Mobilization of uranium in the alluvial plains of Punjab, India: release from sediment to groundwater. *Goldschmidt virtual*. <https://doi.org/10.7185/gold2021.3596>, 2021.
- Banning, A., Pawletko, N., Roder, J., Kubeck, C., Wisotzky, F., 2017. Ex situ groundwater treatment triggering the mobilization of geogenic uranium from aquifer sediments. *Sci. Total Environ.* 587, 371–380. <https://doi.org/10.1016/j.scitotenv.2017.02.162>.
- Baweja, S., Aggarwal, R., Brar, M., Lal, R., 2017. Groundwater depletion in Punjab, India. *Soil Sci.* 3, 1–5. <https://doi.org/10.1081/e-ess3-120052901>.
- Bernhard, G., Geipel, G., Brendler, V., Nitsche, H., 1998. Uranium speciation in waters of different uranium mining areas. *J. Alloys Compd.* 271, 201–205. [https://doi.org/10.1016/S0925-8388\(98\)00054-1](https://doi.org/10.1016/S0925-8388(98)00054-1).
- Bonsor, H.C., MacDonald, A.M., Ahmed, K.M., Burgess, W.G., Basharat, M., Calow, R.C., Dixit, A., Foster, S.S.D., Gopal, K., Lapworth, D.J., Moench, M., 2017. Hydrogeological typologies of the indo-gangetic basin alluvial aquifer, south asia. *Hydrogeol. J.* 25 (5), 1377–1406. <https://doi.org/10.1007/s10040-017-1550-z>.
- Brindha, K., Elango, L., 2013. Occurrence of uranium in groundwater of a shallow granitic aquifer and its suitability for domestic use in southern India. *J. Radioanal. Nucl. Chem.* 295 (1), 357–367. <https://doi.org/10.1007/s10967-012-2090-6>.
- Burow, K.R., Belitz, K., Dubrovsky, N.M., Jurgens, B.C., 2017. Large decadal-scale changes in uranium and bicarbonate in groundwater of the irrigated western. *U.S. Sci. Total Environ.* 586, 87–95. <https://doi.org/10.1016/j.scitotenv.2017.01.220>.
- Cgwb, 2014. *Concept Note on Geogenic Contamination of Groundwater in India*. Central Ground Water Board, Ministry of Water Resources. Govt. of India. Geogenic Final. pdf (cgwb.gov.in).
- Cgwb, 2017a. Report on Aquifer Mapping and Management Plan, Barnala District, Punjab. Central Ground Water Board. Available online: <http://cgwb.gov.in/AQM/NAQUIM-REPORT/Punjab/Barnala.pdf>.
- Cgwb, 2017b. Ground water year book, Punjab and Chandigarh (ut). Central ground water board. Available online: <http://cgwb.gov.in/GW-Year-Book-State.html>.
- Chen, B., Roos, P., Borggaard, O.K., Zhu, Y.G., Jakobsen, I., 2005. Mycorrhiza and root hairs in barley enhance acquisition of phosphorus and uranium from phosphate rock but mycorrhiza decreases root to shoot uranium transfer. *New Phytol.* 165 (2), 591–598. <https://doi.org/10.1111/j.1469-8137.2004.01244.x>.
- Chopra, R.P.S., Krishan, G., 2014. Analysis of aquifer characteristics and groundwater quality in southwest Punjab, India. *J. Earth Sci. Eng.* 4, 597–604. <https://doi.org/10.17265/2159-581X/2014.10.002>.
- Clark, I.D., Fritz, P., 1997. *Environmental Isotopes in Hydrology*. Lewis Publishers, New York.
- Cothren, C.R., Lappenbusch, W.L., 1983. Occurrence of uranium in drinking water in the US. *Health Phys.* 45, 89–99. <https://doi.org/10.1097/00004032-198307000-00009>.
- Coyte, R.M., Jain, R.C., Srivastava, S.K., Sharma, K.C., Khalil, A., Ma, L., Vengosh, A., 2018. Large-scale uranium contamination of groundwater resources in India. *Environ. Sci. Technol. Lett.* 5 (6), 341–347. <https://doi.org/10.1021/acs.estlett.8b00215>.
- Craig, H., 1961. Isotopic variations in meteoric waters. *Sci* 133, 1702–1703. <https://doi.org/10.1126/science.133.3465.1702>.
- Dang, H.S., Jaiswal, D.D., Parameswaran, M., Deodhar, K.P., Krishnamony, S., 1996. Age dependent physical and anatomical Indian data for application in internal dosimetry. *Radiat. Protect. Dosim.* 63 (3), 217–222. <https://doi.org/10.1093/oxfordjournals.rpd.a031532>.
- Dehnavi, A.G., Sarikhani, R., Nagaraju, D., 2011. Hydro geochemical and rock water interaction studies in East of Kurdistan, NW of Iran. *Int. J. Environ. Sci. Res.* 1 (1), 16–22.
- Dhanaraju, R., Sinha, P.K., Umamaheswar, K., Gorikhan, A., 1985. Petrology of the uraniferous Siwalik sandstones from near Loharkar, Himachal Pradesh, India, with a note on the physico chemical conditions of transportation and deposition on uranium. *J. Indian Assoc. Sedimentol.* 5, 12–22.
- Dreeson, D.R., Williams, J.M., Marple, M.L., Gladney, E.S., Perrin, D.R., 1982. Mobility and bioavailability of uranium mill tailings constituents. *Environ. Sci. Technol.* 16, 70209. <https://doi.org/10.1021/es00104a013>.
- Echevarria, G., Sheppard, M.I., Morel, J., 2001. Effect of pH on the sorption of uranium in soils. *J. Environ. Radioact.* 53 (2), 257–264. [https://doi.org/10.1016/S0265-931X\(00\)00116-8](https://doi.org/10.1016/S0265-931X(00)00116-8).
- Elango, L., Kannan, R., 2007. Rock–water interaction and its control on chemical composition of groundwater. *Dev. Environ. Sci.* 5, 229–243. [https://doi.org/10.1016/S1474-8177\(07\)05011-5](https://doi.org/10.1016/S1474-8177(07)05011-5).
- Finch, R., Murakami, T., 1999. Systematics and paragenesis of Uranium minerals. In: Burns, P.C., Finch, R. (Eds.), *Uranium: Mineralogy, Geochemistry and the Environment*, Reviews in Mineralogy, vol. 38, pp. 91–180. <https://doi.org/10.1515/9781501509193-008>.
- Frengstad, B., Skrede, A.K.M., Banks, D., Krog, J.R., Siewers, U., 2000. The chemistry of Norwegian groundwaters: III. The distribution of trace elements in 476 crystalline bedrock groundwaters, as analysed by ICP-MS techniques. *Sci. Total Environ.* 246 (1), 21–40. [https://doi.org/10.1016/S0048-9697\(99\)00413-1](https://doi.org/10.1016/S0048-9697(99)00413-1).
- Gibbs, R.J., 1970. Mechanisms controlling world water chemistry. *Sci* 170 (3962), 1088–1090. <https://doi.org/10.1126/science.170.3962.1088>.
- Hakonson-Hayes, A.C., Fresquez, P.R., Whicker, F.W., 2002. Assessing potential risks from exposure to natural uranium in well water. *J. Environ. Radioact.* 59 (1), 29–40. [https://doi.org/10.1016/S0265-931X\(01\)00034-0](https://doi.org/10.1016/S0265-931X(01)00034-0).
- HDR, 2009. Human Development Report. National Resource Centre for Urban Poverty and All India Institute of Local Self Government, Mumbai, India.
- Hundal, H.S., 2011. Geochemistry and assessment of hydrogeochemical processes in groundwater in the southern part of Bathinda district of Punjab, northwest India. *Environ. Earth Sci.* 64 (7), 1823–1833.
- ICRP, 2008. The 2007 recommendations of the international commission on radiological protection. *ICRP publication 103. Ann. ICRP* 37 (2–4), 1–332.
- Jain, S.C., Metha, S.C., Kumar, B., Reddy, A.R., Nagaratnam, A., 1995. Formulation of the reference Indian adult: anatomic and physiologic data. *Health Phys.* 68 (4), 509–522. <https://doi.org/10.1097/00004032-199504000-00008>.
- Jerden, J.L., Sinha, A.K., 2006. Geochemical coupling of uranium and phosphorus in soils overlying an unmined uranium deposit: coles Hill, Virginia. *J. Geochem. Explor.* 91 (1–3), 56–70. <https://doi.org/10.1016/j.gexplo.2005.12.003>.
- Jurgens, B.C., Burow, K.R., Dalgish, B.A., Shelton, J.L., 2008. Hydrogeology, Water Chemistry, and Factors Affecting the Transport of Contaminants in the Zone of Contribution of a Public-Supply Well in Modesto, Eastern San Joaquin Valley, California. U.S. Geological Survey Scientific Investigations. Report 2008-5156. <http://pubs.er.usgs.gov/publication/sir20085156>.
- Jurgens, B.C., Fram, M.S., Belitz, K., Burow, K.R., Landon, M.K., 2010. Effects of groundwater development on uranium: central valley, California, USA. *Ground Water* 48 (6), 913–928. <https://doi.org/10.1111/j.1745-6584.2009.00635.x>.
- Kar, A., 1999. A Hitherto Unknown Palaeodrainage System from Radar Imagery of Southeastern that Desert and its Significance. *Mem. Geol. Soc. India*, pp. 229–236.
- Kaul, R., Umamaheshwar, K., Chandrasekaran, S., Deshmukh, R.D., Swarnkar, B.M., 1993. Uranium mineralization in the siwaliks of northwestern himalaya, India. *J. Geol. Soc. India* 9, 773–785.
- Keesari, T., Sharma, D.A., Rishi, M.S., Pant, D., Mohokar, H.V., Jaryal, A.K., Sinha, U.K., 2017. Isotope investigation on groundwater recharge and dynamics in shallow and deep alluvial aquifers of southwest Punjab. *Appl. Radiat. Isot.* 129, 163–170. <https://doi.org/10.1016/j.apradiso.2017.07.022>.
- Kendall, C., Caldwell, E.A., 1998. Fundamentals of isotope geochemistry. In: *Isotope Tracers in Catchment Hydrology*. Elsevier, pp. 51–86. <https://doi.org/10.1016/B978-0-444-81546-0.50009-4>.
- Krestou, A., Panias, D., 2004. Uranium (VI) speciation diagrams in the UO₂ 2+ / CO₃ 2- / H₂ O system at 25 °C. *Eur. J. Miner. Process. Environ. Protect.* 4 (2).
- Krishan, G., Kumar, B., Sudarsan, N., Rao, M.S., Ghosh, N.C., Taloor, A.K., Bhattacharya, P., Singh, S., Kumar, C.P., Sharma, A., Jain, S.K., Sidhu, B.S., Kumar, S., Vasisth, R., 2021. Isotopes (δ18O, δD and 3H) variations in groundwater with emphasis on salinization in the state of Punjab, India. *Sci. Total Environ.* 789, 148051. <https://doi.org/10.1016/j.scitotenv.2021.148051>.
- Krupa, S.L., Belanger, T.V., Heck, H.H., Brock, J.T., Jones, B.J., 1998. Krupaseep- the next generation seepage meter. *J. Coast Res.* 210–213.
- Krynine, P.D., 1937. Petrography and genesis of the Siwalik series. *Am. J. Sci.* 5 (34), 422–446. <https://doi.org/10.2475/ajs.s5-34.204.422>.
- Kumar, A., Rout, S., Narayanan, U., Mishra, M.K., 2011. Geochemical modelling of Uranium speciation in the subsurface aquatic environment of Punjab state in India. *J. Geol. Min. Res.* 3 (5), 137–146.
- Kumar, A., Tripathi, R.M., Rout, S., Mishra, M.K., Ravi, P.M., Ghosh, A.K., 2014. Characterization of groundwater composition in Punjab state with special emphasis on uranium content, speciation and mobility. *Radiochim. Acta* 102 (3), 239–254. <https://doi.org/10.1515/ract-2014-2109>.
- Kumar, R., Vaid, U., Mittal, S., 2018. Water Crisis: Issues and Challenges in Punjab, vols. 93–103. *Water Resour. Manag.* https://doi.org/10.1007/978-981-10-5711-3_7.
- Kumar, R., Mittal, S., Sahoo, P.K., Sahoo, S.K., 2021. Source apportionment, chemometric pattern recognition and health risk assessment of groundwater from southwestern Punjab, India. *Environ. Geochem. Health* 43 (2), 733–755. <https://doi.org/10.1007/s10653-020-00518-1>.
- Kurtito, P., Harmoinen, A., Saha, H., Salonen, L., Karpas, Z., Komulainen, H., Auvinen, A., 2006. Kidney toxicity of ingested uranium from drinking water. *Am. J. Kidney Dis.* 47 (6), 972–982. <https://doi.org/10.1053/j.ajkd.2006.03.002>.
- Langmuir, D., 1978. Uranium solution-mineral equilibria at low temperatures with applications to sedimentary ore deposits. *Geochem. Cosmochim. Acta* 42, 547–569. <https://doi.org/10.2172/6774931>.
- Langmuir, D., 1997. *Aqueous Environmental Geochemistry*. Prentice Hall, Upper Saddle River, NJ, 600.
- Li, S., Wang, X., Huang, Z., Du, L., Tan, Z., Fu, Y., Wang, X., 2016. Sorption and desorption of uranium (VI) on GMZ bentonite: effect of pH, ionic strength, foreign ions and humic substances. *J. Radioanal. Nucl. Chem.* 308 (3), 877–886. <https://doi.org/10.1007/s10967-015-4513-7>.
- MacDonald, A.M., Bonsor, H.C., Ahmed, K.M., Burgess, W.G., Basharat, M., Calow, R.C., Dixit, A., Foster, S.S.D., Gopal, K., Lapworth, D.J., Lark, R.M., 2016. Groundwater quality and depletion in the Indo-Gangetic Basin mapped from in situ observations. *Nat. Geosci.* 9 (10), 762–766. <https://doi.org/10.1038/ngeo2791>.

- Maheer, K., Bargar, J.R., Brown, G.E., 2013. Environmental speciation of actinides. *Inorg. Chem.* 52 (7), 3510–3532. <https://doi.org/10.1021/ic301686d>.
- Moss, M., McCurdy, R., Dooley, K., Givener, M., Dymond, L., Slater, J.M., Courneya, M., 1983. Uranium in drinking water - report on clinical studies in Nova Scotia. In: Savory, S.S. Brown & E.J. (Ed.), *Chemical Toxicology and Clinical Chemistry of Metals*. Proceedings of 2. International Conference Held in Montreal, Canada, 19-22 July 1983. https://inis.iaea.org/search/search.aspx?orig_q=RN:16006189.
- Nair, S., Karimzadeh, L., Merkel, B.J., 2014. Sorption of uranyl and arsenate on SiO_2 , Al_2O_3 , TiO_2 and FeOOH . *Environ. Earth Sci.* 72 (9), 3507–3512. <https://doi.org/10.1007/s12665-014-3258-x>.
- National Remote Sensing Centre, 2015-16. Land use land cover map, Punjab (50K). Bhuvan, geo-platform of Indian space Research organization (ISRO). <https://bhuvan-app1.nrsc.gov.in/thematic/thematic/index.php>.
- Négrel, P., Petelet-Giraud, E., Barbier, J., Gautier, E., 2003. Surface water-groundwater interactions in an alluvial plain: chemical and isotopic systematics. *J. Hydrol.* 277, 248–267. [https://doi.org/10.1016/S0022-1694\(03\)00125-2](https://doi.org/10.1016/S0022-1694(03)00125-2).
- Nriagu, J., Nam, D.H., Ayanwola, T.A., Dinh, H., Erdenechimeg, E., Ochir, C., Bolomaa, T.A., 2012. High levels of uranium in groundwater of Ulaanbaatar, Mongolia. *Sci. Total Environ.* 414, 722–726. <https://doi.org/10.1016/j.scitotenv.2011.11.037>.
- Pant, D., Keesari, T., Sharma, D., Rishi, M., Singh, G., Jaryal, A., Sinha, U.K., Dash, A., Tripathi, R.M., 2017. Study on uranium contamination in groundwater of Faridkot and Muktsar districts of Punjab using stable isotopes of water. *J. Radioanal. Nucl. Chem.* 313 (3), 635–645. <https://doi.org/10.1007/s10967-017-5284-0>.
- Pant, D., Keesari, T., Roy, A., Sinha, U.K., Singh, M., Jain, S.K., Tripathi, R.M., 2019. Study on groundwater quality in parts of Rajasthan with special reference to uranium contamination. *J. Radioanal. Nucl. Chem.* 322 (1), 165–171. <https://doi.org/10.1007/s10967-019-06525-6>.
- Patnaik, R., Lahiri, S., Chahar, V., Naskar, N., Sharma, P.K., Avhad, D.K., Bassan, M.K.T., Knolle, F., Schnug, E., Srivastava, A., 2016. Study of uranium mobilization from Himalayan siwaliks to the Malwa region of Punjab state in India. *J. Radioanal. Nucl. Chem.* 308, 913–918. <https://doi.org/10.1007/s10967-015-4578-3>.
- Paul, R.K., Paul, A.K., Gurung, B., Bithal, P., 2015. Temperature trend in different agro-climatic zones in India. *Mausam* 66 (4), 841–846.
- PC, 2013. Report of the High Level Expert Group on Water Logging in Punjab. Government of India, Planning Commission. https://niti.gov.in/planningcommission.gov.in/docs/reports/genrep/rep_waterpunjab2702.pdf.
- Phadke, A.V., Mahadevan, T.M., Narayan, D., Saraswat, A.C., 1985. Uranium Mineralisation in Some Phanerozoic Sandstones in India. IAEA Technical Document No. p. 328.
- Piper, A.M., 1944. A graphic procedure in the geochemical interpretation of water-analyses. *Eos. Trans. Am. Geophys. Union* 25 (6), 914–928. <https://doi.org/10.1029/TR025i006p00914>.
- Porcelli, D., Swarzenski, P.W., 2003. The behavior of U- and Th-series nuclides in groundwater. *Rev. Mineral. Geochem.* 52 (1), 317–361. <https://doi.org/10.1515/9781501509308-013>.
- Rai, S.P., Joshi, S.K., Sinha, R., Gupta, S., Densmore, A.L., Rawat, Y.S., Shekhar, S., 2014. Sources and Relative Timing of Groundwater Recharge in Northwest India Using Environmental Isotopes. American Geophysical Union, Fall Meeting, 2014, abstract #H43N-1170.
- Rajmohan, N., Elango, L., 2003. Identification and evolution of hydrogeochemical processes in the groundwater environment in an area of the Palar and Cheyyar River Basins, Southern India. *Environ. Geol.* 46 (1), 47–61. <https://doi.org/10.1007/s00254-004-1012-5>.
- Rao, M.S., Krishan, G., Kumar, C.P., Purushothaman, P., Kumar, S., 2017. Observing changes in groundwater resource using hydro-chemical and isotopic parameters: a case study from Bist Doab, Punjab. *Environ. Earth Sci.* 76, 175. <https://doi.org/10.1007/s12665-017-6492-1>.
- Rishi, M.S., Keesari, T., Sharma, D.A., Pant, D., Sinha, U.K., 2017. Spatial trends in uranium distribution in groundwaters of Southwest Punjab, India - a hydrochemical perspective. *J. Radioanal. Nucl. Chem.* 311 (3), 1937–1945. <https://doi.org/10.1007/s10967-017-5178-1>.
- Rojas, J.L., 1989. Introduction to in Situ Leaching of Uranium: Technical, Environmental and Economic Aspects. IAEA Technical Document No. 492.
- Sahoo, S., Swain, S., Goswami, A., Sharma, R., Pateriya, B., 2021. Assessment of trends and multi-decadal changes in groundwater level in parts of the Malwa region, Punjab, India. *Groundw. Sustain. Dev.* 14, 100644. <https://doi.org/10.1016/j.gsd.2021.100644>.
- Saini, K., Singh, P., Bajwa, B.S., 2016. Comparative statistical analysis of carcinogenic and non-carcinogenic effects of uranium in groundwater samples from different regions of Punjab, India. *Appl. Radiat. Isot.* 118, 196–202. <https://doi.org/10.1016/j.apradiso.2016.09.014>.
- Sarin, M.M., Krishnaswami, S., Dilli, K., Somayajulu, B.L.K., Moore, W.S., 1989. Major ion chemistry of the Ganga-Brahmaputra river system: weathering processes and fluxes to the Bay of Bengal. *Geochim. Cosmochim. Acta* 53 (5), 997–1009. [https://doi.org/10.1016/0016-7037\(89\)90205-6](https://doi.org/10.1016/0016-7037(89)90205-6).
- Schoeller, H., 1977. *Geochemistry of Groundwater*. Groundwater Studies, an International Guide for Research and Practice. UNESCO, Paris, pp. 1–18.
- Sharma, D.A., Rishi, M.S., Keesari, T., Pant, D., Singh, R., Thakur, N., Sinha, U.K., 2017. Distribution of uranium in groundwaters of Bathinda and Mansa districts of Punjab, India: inferences from an isotope hydrochemical study. *J. Radioanal. Nucl. Chem.* 313 (3), 625–633. <https://doi.org/10.1007/s10967-017-5288-9>.
- Sharma, D.A., Keesari, T., Rishi, M.S., Pant, D., 2018. A study on the role of hydrogeology on the distribution of uranium in alluvial aquifers of northwest India. *Environ. Monit. Assess.* 190 (12), 746. <https://doi.org/10.1007/s10661-018-7112-6>.
- Sharma, T., Litoria, P.K., Bajwa, B.S., Kaur, I., 2021. Appraisal of groundwater quality and associated risks in Mansa district (Punjab, India). *Environ. Monit. Assess.* 193 (4), 1–21. <https://doi.org/10.1007/s10661-021-08892-8>.
- Sidhu, B.S., Sharda, R., Singh, S., 2021. Spatio-temporal assessment of groundwater depletion in Punjab, India. *Groundw. Sustain. Dev.* 12, 100498. <https://doi.org/10.1016/j.gsd.2020.100498>.
- Singh, K.P., Kishore, N.K., 2010. Uranium in groundwater. Some observations from parts of Talwandi Sabo block, district Bathinda, Punjab. *Proceedings Punjab Science Congress*. Feb 7-9.
- Singh, J., Singh, L., Singh, S., 1995. High U-contents observed in some drinking water of Punjab, India. *J. Environ. Radioact.* 26, 217–222. [https://doi.org/10.1016/0265-931x\(94\)00037-w](https://doi.org/10.1016/0265-931x(94)00037-w).
- Singh, H., Singh, J., Singh, S., Bajwa, B.S., 2009. Uranium concentration in drinking water samples using SSNTDs. *Indian J. Phys.* 83 (7), 1039–1044. <https://doi.org/10.1007/s12648-009-0065-4>.
- Singh, C.K., Kumar, A., Shashtri, S., Kumar, A., Kumar, P., Mallick, J., 2017a. Multivariate statistical analysis and geochemical modeling for geochemical assessment of groundwater of Delhi, India. *J. Geochem. Explor.* 175, 59–71. <https://doi.org/10.1016/j.gexplo.2017.01.001>.
- Singh, S., Singh, T., Deepthi, S.S., Kaur, A., Mahajan, S., Lal, M., 2017b. Major sites of cancer occurrence among men in Amritsar District, India: findings from a tertiary care. *Hindu* 87, 37–66.
- Smedley, P.L., Smith, B., Abesser, C., Lapworth, D., 2006. *Uranium Occurrence and Behaviour in British Groundwater*. British Geological Survey Groundwater Systems & Water Quality Programme Commissioned Report CR/06/050.
- Spalding, R.F., Sackett, W.M., 1972. Uranium in runoff from the Gulf of Mexico distributive province: anomalous concentrations. *Sci* 175, 62931. <https://doi.org/10.1126/science.175.4022.629>.
- Szecsody, J.E., Krupka, K.M., Williams, M.D., Cantrell, K.J., Resch, C.T., Fruchter, J.S., 1998. Uranium Mobility during in Situ Redox Manipulation of the 100 Areas of the Hanford Site (No. PNNL-12048). Pacific Northwest National Lab.(PNNL), Richland, WA (United States). <https://doi.org/10.2172/2120>.
- Tadmor, J., 1986. Radioactivity from coal-fired power plants: a review. *J. Environ. Radioact.* 4, 177–204. [https://doi.org/10.1016/0265-931x\(86\)90010-x](https://doi.org/10.1016/0265-931x(86)90010-x).
- Tarki, M., Hammadi, M.B., El Mejri, H., Dassi, L., 2016. Assessment of hydrochemical processes and groundwater hydrodynamics in a multilayer aquifer system under long-term irrigation condition: a case study of Nezaoua basin, southern Tunisia. *Appl. Radiat. Isot.* 110, 138–149. <https://doi.org/10.1016/j.apradiso.2016.01.009>.
- Thivya, C., Chidambaram, S., Keesari, T., Prasanna, M.V., Thilagavathi, R., Adithya, V.S., Singaraja, C., 2016. Lithological and hydrochemical controls on distribution and speciation of uranium in groundwaters of hard-rock granitic aquifers of Madurai District, Tamil Nadu (India). *Environ. Geochem. Health* 38 (2), 497–509. <https://doi.org/10.1007/s10653-015-9735-7>.
- Tripathi, R.M., Sahoo, S.K., Mohapatra, S., Lenka, P., Dubey, J.S., Puranik, V.D., 2013. Study of uranium isotopic composition in groundwater and deviation from secular equilibrium condition. *J. Radioanal. Nucl. Chem.* 295, 1195–1200. <https://doi.org/10.1007/s10967-012-1992-7>.
- Tripathi, A., Mishra, A.K., Verma, G., 2016. Impact of preservation of subsoil water act on groundwater depletion: the case of Punjab, India. *Environ. Manag.* 58 (1), 48–59. <https://doi.org/10.1007/s00267-016-0693-3>.
- Trivedy, R.K., Goel, P.K., 1984. *Chemical and Biological Methods for Water Pollution Studies*. Environmental publications.
- USEPA, 1991. Review of RSC (Relative Source Contribution) Analysis. Report prepared by Wade Miller Associates, Inc. for the office of drinking water, May 9 (1991). US Environmental Protection Agency.
- USEPA, 2000. National Primary Drinking Water Regulation; Radionuclides; Final Rule. US Environmental Protection Agency, Washington, DC.
- USEPA, 2005. Human Health Risk Assessment Protocol for Hazardous Waste Combustion Facilities. EPA530-R-05-006. US Environmental Protection Agency, Washington, DC.
- Virk, H.S., 2019. Uranium content anomalies in groundwater of Barnala district of malwa belt of Punjab (India) for the assessment of excess cancer risk. *Res. Rev.: J. Oncol. Hematol.* 8 (1), 19–26. <https://doi.org/10.37591/rjoo.v8i1.718>.
- Welch, A.H., Lico, M.S., 1998. Factors controlling As and U in shallow ground water, southern Carson Desert, Nevada. *Appl. Geochem.* 13, 521–539. [https://doi.org/10.1016/S0883-2927\(97\)00083-8](https://doi.org/10.1016/S0883-2927(97)00083-8).
- Who, 2011a–2018. World Health Organization. Guidelines for Drinking-Water Quality, fourth ed. Available online: https://apps.who.int/iris/bitstream/handle/10665/44584/9789241548151_eng.pdf;jsessionid=867B008DE40ACC534C919B4AC3509C87?sequence=1.
- Who, 2011b. *World Health Statistics*. World Health Organization, Geneva.
- Wrenn, M.E., Durbin, P.W., Howard, B., Lipsztein, J., Rundo, J., Still, E.T., Willis, D.L., 1985. Metabolism of ingested U and RA. *Health Phys.* 48 (5), 601–633. <https://doi.org/10.1097/00004032-198505000-00004>.
- Wu, Y., Wang, Y., Xie, X., 2014. Occurrence, behavior and distribution of high levels of uranium in shallow groundwater at Datong basin, northern China. *Sci. Total Environ.* 472, 809–817. <https://doi.org/10.1016/j.scitotenv.2013.11.109>.
- Wu, Y., Li, J., Wang, Y., Xie, X., 2018. Variations of uranium concentrations in a multi-aquifer system under the impact of surface water-groundwater interaction. *J. Contam. Hydrol.* 211, 65–76. <https://doi.org/10.1016/j.jconhyd.2018.03.007>.